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THE SYSTEM $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$

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ABSTRACT

The thermal-equilibrium relationships in the system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$ have been experimentally investigated by the quenching method. The data are tabulated, and the results are presented in equilibrium diagrams. Complex solid solutions in all the crystalline phases render the system nonternary throughout, and courses of crystallization must be viewed in terms of the quaternary system of the four component oxides. The composition of gehlenite mix-crystals obtained in this investigation is best expressed by assuming solid solution with a hitherto unknown "molecule," of the composition $CaO \cdot 2Al_2O_3$.

INTRODUCTION

This investigation is concerned with the thermal-equilibrium relationships, at atmospheric pressure, of the three silicates, $CaAl_2Si_2O_8$, $Ca_2Al_2SiO_7$, and $NaAlSiO_4$. These compounds are the principal "molecules" of the natural minerals, anorthite, gehlenite, and nepheline, respectively. Anorthite, as an end-member of the plagioclase feldspars, is of prime importance in igneous rocks. Nepheline, a feldspathoid, is the most characteristic mineral of the alkaline igneous rocks, while melilites, of which gehlenite is an end-member, are also feldspathoids that are present in alkaline rocks which are rich in lime. Nepheline and melilite are found associated in some igneous rocks, and gehlenite also occurs as a metamorphic mineral.

The only compound involved in this study that exhibits polymorphism is $NaAlSiO_4$; but carnegieite, the high-temperature modification, has never been found in nature. In addition to the compounds mentioned, the substance β- alumina was encountered and is of petrologic interest, inasmuch as corundum is a typical accessory mineral of the alkaline syenites.

The system constitutes a portion of a plane within the quaternary system soda-lime-alumina-silica. This quaternary system is of major importance in the physicochemical aspects of igneous petrology, as the constituent oxides form more than 80 per cent of the average oxide composition of igneous rocks. The relation of the system studied to the general quaternary system is shown in Figure 1. The system anorthite-gehlenite-nepheline forms one face of a subsidiary tetrahedron anorthite-gehlenite-wollastonite-nepheline within the quaternary system; and the plane anorthite-nepheline-wollastonite is common to a second subsidiary tetrahedron, anorthite-nepheline-wollastonite-albite.*

Forty-six mixtures were prepared and subjected to thermal study. Three of

these compositions were in the plane of anorthite-gehlenite-nepheline + 10 per cent wollastonite, being used to locate the position of the boundary between the fields of anorthite, melilite, and nepheline as it passes through this plane of 10 per cent $CaSiO_3$ in the subsidiary tetrahedron. In addition, five mixtures measured with a platinum-platinum 90 rhodium 10 thermoelement, calibrated at the melting-points of lithium metasilicate (1,201° C.) and diopside (1,391.5° C.). The results of quenching runs were examined microscopically to determine the nature of the phases present at equilibrium. The significant data

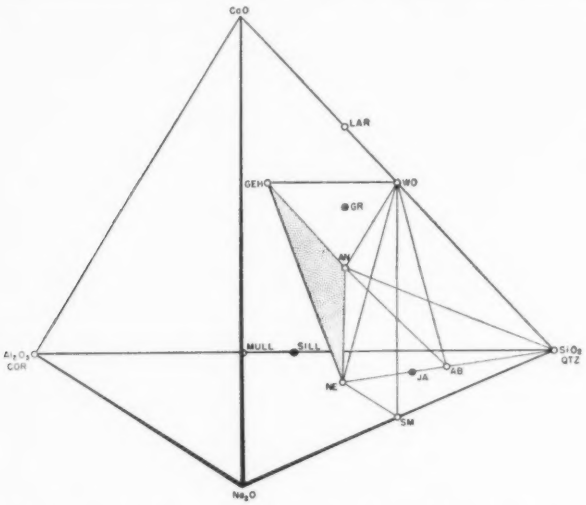

FIG. 1.—The soda-lime-alumina-silica tetrahedron. The shaded area indicates the position of the system anorthite-gehlenite-nepheline within the tetrahedron. Other internal systems that have been investigated are shown by full lines joining the composition points. The plane nepheline-gehlenite-wollastonite plus 10 per cent anorthite has also been investigated (unpublished data of H. S. Yoder). The compounds that have been prepared synthetically are shown as ○; those that occur in nature but that have not been synthesized are shown thus: ⊗.

The abbreviations signify: *AN*, anorthite; *GEH*, gehlenite; *NE*, nepheline; *WO*, wollastonite; *AB*, albite; *JA*, jadeite; *SM*, sodium metasilicate; *SILL*, sillimanite; *MULL*, mullite; *LAR*, larnite; *GR*, grossularite; *QTZ*, quartz; *COR*, corundum.

prepared and examined by H. S. Yoder[1] that fall in the plane AN-GEH-NE are incorporated with the results here presented.

The mixtures were studied as to thermal behavior by means of the quenching method developed in the Geophysical Laboratory of the Carnegie Institution of Washington.[2] Temperatures were are presented in Tables 1 and 2, and from these data the equilibrium diagram has been prepared.

PROPERTIES OF THE CRYSTALLINE PHASES

$CaAl_2Si_2O_8$

Pure $CaAl_2Si_2O_8$ has but one modification—anorthite—which melts at

[1] Unpublished data on the system nepheline-gehlenite-wollastonite plus 10 per cent anorthite.

[2] E. S. Shepherd, G. A. Rankin, and F. E. Wright, "The Binary Systems of Alumina with Silica, Lime, and Magnesia," *Amer. Jour. Sci.*, Vol. XXVIII (4th ser., 1909), p. 308.

1,552° C. It is triclinic, with $\alpha = 1.5755$, $\beta = 1.5832$, and $\gamma = 1.5885$. Crystals obtained in quenching runs are commonly platy, with 010 showing the strongest development. Albite twinning is common. The crystals encountered in this investigation are plagioclase, of a composition not far from pure anorthite, as shown by maximum index values approaching 1.588.

$Ca_2Al_2SiO_7$

Artificial gehlenite also melts congruently, at a temperature of 1,590° C.[3] It is tetragonal, as tabular or short prismatic crystals. Its indices of refraction are $\omega = 1.669$ and $\epsilon = 1.658$. The melilites encountered in this study were generally tabular, thus showing positive elongation on a negative crystal. Globular to shapeless forms were also encountered, particularly in compositions close to a boundary curve. The indices of melilites forming in this system were significantly lower than those of pure gehlenite. Although the indices were accurately determined in but several runs, values as low as $\omega = 1.651$ and $\epsilon' = 1.641$ were encountered. This will be discussed in more detail in a later section concerned with substances in solid solution.

$NaAlSiO_4$

Carnegieite, the high-temperature modification of $NaAlSiO_4$, exists in three forms, all of which are known only from synthetic melts. The highest-temperature form, which is isometric, melts at 1,526° C.[4] and inverts to a triclinic form at 690° C.[5] A second inversion, accompanied by a marked change in birefringence, occurs at 226.5° C. The indices of refraction of the low-temperature form are given by Bowen[6] as $\alpha = 1.514$, $\beta = 1.514$, $\gamma = 1.509$. Carnegieite usually occurs in globular masses, showing no external crystal form and, typically, has intricate polysynthetic twinning that forms as a result of inversion.

Pure artificial nepheline inverts to carnegieite at a temperature of 1,254 ± 5° C.[7] Bowen[8] found that anorthite, which may go into solid solution with nepheline to the extent of 35 per cent, raises the inversion temperature 100° C., while albite in solid solution brings about a rise of 30° at its maximum concentration.[9] Smalley, in investigating the system $NaAlSiO_4$–$Ca_2Al_2SiO_7$, found an inversion range extending from about 50° above, to 100° below, the normal inversion point.[10] However, the nonbinary nature of the system precluded making any assured statement as to the exact nature of the solid solution.

Nepheline is hexagonal and crystallizes from melts as prismatic crystals with basal pinacoid. The indices as determined by Bowen on the pure synthetic material are $\epsilon = 1.533$ and $\omega = 1.537$.[11] He found that the indices of the limiting anorthite-nepheline solid solution are

[3] E. F. Osborn and J. F. Schairer, "The Ternary System, Pseudo-wollastonite-Akermanite-Gehlenite," *Amer. Jour. Sci.*, Vol. CCXXXIX (1941), p. 719.

[4] N. L. Bowen, "The Binary System: $Na_2Al_2Si_2O_8$ (Nephelite, Carnegieite)–$CaAl_2Si_2O_8$ (Anorthite)," *Amer. Jour. Sci.*, Vol. XXXIII (4th ser., 1912), pp. 554 ff.

[5] N. L. Bowen and J. W. Greig, "The Crystalline Modifications of $NaAlSiO_4$," *Amer. Jour. Sci.*, Vol. X (5th ser., 1925), pp. 208–10.

[6] P. 564 of ftn. 4.

[7] J. W. Greig and Tom. F. W. Barth, "The System $Na_2O \cdot Al_2O_3 \cdot 2SiO_2$ (Nephelite, Carnegieite)–$Na_2O \cdot Al_2O_3 \cdot 6SiO_2$ (Albite)," *Amer. Jour. Sci.*, Vol. XXXV+A (5th ser., 1938) p. 108.

[8] P. 571 of ftn. 4.

[9] P. 109 of ftn. 7.

[10] Robert G. Smalley, "The System $NaAlSiO_4$–$Ca_2Al_2SiO_7$," *Jour. Geol.*, Vol. LV, No. 1 (January, 1947), p. 28.

[11] P. 566 of ftn. 4.

$\epsilon = 1.539$, $\omega = 1.537$, while solid solution of albite in nepheline has been found to lower the indices of the latter somewhat.[12] Unusually high indices were noted by Smalley in nephelines crystallizing from the system $NaAlSiO_4$–$Ca_2Al_2SiO_7$, values as high as 1.552 being reached in crystals from melts having 35 per cent gehlenite.[13] The present study indicates that the solid solution responsible for this rise remains effective upon addition of $CaAl_2Si_2O_8$, as values of $\omega = 1.548$ and $\epsilon > 1.548$ were obtained from runs at a composition of approximately 35 per cent nepheline, 20 per cent gehlenite, and 45 per cent anorthite.

β–Al_2O_3

The β–Al_2O_3 here encountered occurs in thin hexagonal plates, exhibiting high relief and high birefringence when viewed on edge. Parallel extinction and positive elongation are shown, the sign of elongation coupled with the platy habit indicating that the crystals are optically negative. No indices were determined, but G. A. Rankin and H. E. Merwin gave the values $\omega = 1.677 \pm .003$ and $\epsilon = 1.635$–1.650 for the β–Al_2O_3 found in preparations thought to be pure alumina.[14] Rankin and Merwin determined the melting-point as 2,050° ± 20° C.

The exact relations of β–Al_2O_3 to the other known forms of Al_2O_3 are not completely understood. The indices are considerably lower than those of corundum, and chemical and X-ray analyses indicate that soda is present in the structure. On the basis of X-ray work the formula $Na_2O \cdot 11Al_2O_3$ was proposed by C. A. Beevers and M. A. S. Ross,[15] whereas W. L. Bragg, C. Gottfried, and J. West had somewhat earlier determined it as $\frac{1}{2}\ Na_2O \cdot 11\frac{1}{2}Al_2O_3$.[16]

Recent work at the Geophysical Laboratory of the Carnegie Institution of Washington has shown that β–Al_2O_3 is converted to corundum when held at elevated temperatures for long periods,[17] indicating that β–Al_2O_3 is metastable. The melting-point as determined by Rankin and Merwin is almost exactly that of pure α–Al_2O_3, so it is likely that their crystals had undergone the conversion to corundum before melting. It is possible that β–Al_2O_3 is an example of a "stuffed lattice," in which Na atoms assume random interstitial positions in the network. Certain soda-bearing tridymites have been interpreted in this way by M. J. Buerger.[18] This explanation would account for the disagreements of different investigators, as it does not require any fixed ratio of soda to alumina. It is also consistent with the findings of the Geophysical Laboratory, as the Na atoms, given sufficient time, might tend to be "squeezed out" of the structure.

PREVIOUS INVESTIGATIONS

THE SYSTEM $CaAl_2Si_2O_8$–$NaAlSiO_4$

The system $CaAl_2Si_2O_8$–$NaAlSiO_4$ was investigated by Bowen,[19] and his equilibrium diagram is shown in Figure 2, with modifications based on work of

[12] W. R. Foster, "The System $NaAlSi_3O_8$–$CaSiO_3$–$NaAlSiO_4$," *Jour. Geol.*, Vol. L (1942), p. 157.

[13] P. 28 of ftn. 10.

[14] "The Ternary System CaO–Al_2O_3–MgO," *Jour. Amer. Chem. Soc.*, Vol. XXXVIII (1916), pp. 566–88.

[15] "The Crystal Structure of β–Al_2O_3," *Zeitschr. f. Kryst. u. Min.*, Vol. XCVII (1937), pp. 59–66.

[16] "The Structure of β–Al_2O_3," *Zeitschr. f. Krysl. u. Min.*, Vol. LXXVII (1931), pp. 255–74.

[17] J. F. Schairer and N. L. Bowen, "Melting Relations in the Systems Na_2O–Al_2O_3–SiO_2 and K_2O–Al_2O_3–SiO_2," *Amer. Jour. Sci.*, Vol. CCXLV (April, 1947) pp. 193–204.

[18] *Science*, Vol. XCV (1942), pp. 20–21.

[19] Pp. 551–73 of ftn. 4.

J. F. Schairer[20] and of W. K. Gummer.[21] This system exhibits partial solid solution between the two end-members, complicated by the carnegieite-nepheline inversion. It was, in fact, the first-found example in silicates of the combined phenomena of solid solution and enantiotropism theoretically proposed by Roozeboom. The eutectic was placed at 46 per cent anorthite by Bowen and at a temperature of 1,304° C. The inversion takes place at 1,352° C., a rise of 104° above the inversion in pure $NaAlSiO_4$, as a result of the solid solution of anorthite in nepheline to the extent of 35 weight per cent. Carnegieite takes but approximately 5 per cent anorthite in solid solution.

Bowen encountered "alumina" in some of his mixtures and accounted for it by soda volatilization at the high temperatures required for fusion of the glasses. This actually occurs but is only part of the story. The β-form was at that time unknown, but, in investigating the system $NaAlSiO_4$–$CaAl_2Si_2O_8$–SiO_2, Schairer[22] found β–Al_2O_3 in mixtures adjacent to the nepheline-anorthite join; and, on preparing mixtures along that join, he also found a considerable range in which β–Al_2O_3 is the primary phase. This work was somewhat amplified by Gummer,[23] who determined the extent of the field of β–Al_2O_3 in the system $CaSiO_3$–$CaAl_2Si_2O_8$–$NaAlSiO_4$. The liquidus for β–Al_2O_3 has not been definitely established as yet, but its outline, based on the evidence on hand, is shown by a dashed line on the diagram.

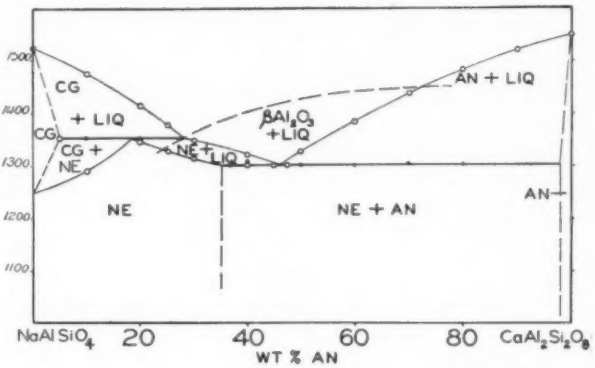

FIG. 2.—Equilibrium diagram for the system $CaAl_2Si_2O_8$–$NaAlSiO_4$ (after Bowen, with modifications after work by Schairer and Gummer).

This intrusion of a field of β–Al_2O_3 upsets the binary nature of the system. The field of nepheline plus liquid is actually a field of β–Al_2O_3, nepheline, and liquid; and much of the field of anorthite plus liquid, as determined by Bowen, is also a ternary field of β–Al_2O_3, anorthite, and liquid. The subliquidus extent of the field of β–Al_2O_3 has not been determined, but Gummer's work indicates that it is present in several mixtures at 1,200° C.

[20] Unpublished data.

[21] "The System $CaSiO_3$–$CaAl_2Si_2O_8$–$NaAlSiO_4$," *Jour. Geol.*, Vol. LI, No. 8 (November–December, 1943), pp. 508–9.

[22] Unpublished data.

[23] Pp. 508–9 of ftn. 21.

THE SYSTEM $NaAlSiO_4$–$Ca_2Al_2SiO_7$

The equilibrium diagram of this system is presented in Figure 3, as determined by Smalley.[24] The unusual nature of this system merits some discussion, which is here presented largely as taken from Smalley's work. In his words: "Even a casual inspection of the equilibrium diagram will suffice to show that this is not a true binary system."[25]

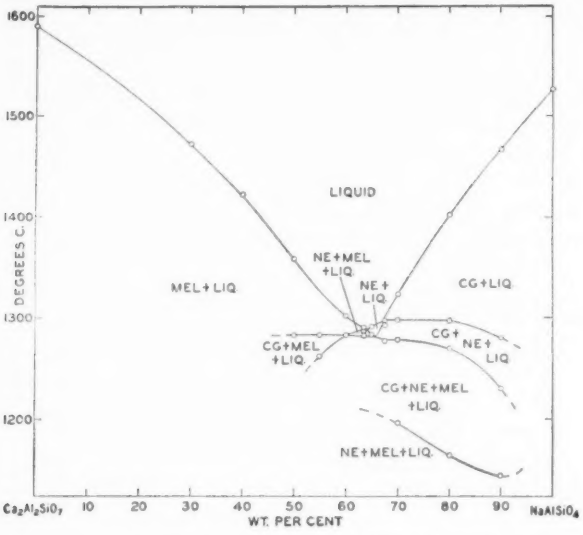

FIG. 3.—Equilibrium diagram for the system $NaAlSiO_4$–$Ca_2Al_2SiO_7$ (after Smalley)

Variations in optical properties of all three of the crystalline phases are indicative of solid solutions; that the solid solutions cannot be expressed in terms of the end-members is obvious from the diagram. The lack of a binary eutectic, of simple solidus curves and the persistence of the liquid phase down to the lowest temperatures of observation, as well as the previously mentioned large temperature range of inversion on both sides of the pseudoeutectic composition, all attest to the nonbinary nature of the system. The inversion relations are further complicated by the unique occurrence, near the pseudoeutectic, of nepheline in equilibrium with liquid at higher temperatures than is carnegieite.

In several fields of this system three phases occur in equilibrium, and in one field four phases are present. Consideration of the phase rule, $P + F = C + 1$, as applied to systems in which the vapor phase is negligible, indicates that in a binary system a maximum of three phases can coexist in equilibrium, and then only at a fixed temperature. Thus the existence of four phases within a temperature composition range requires four components—the four constituent oxides. The join $NaAlSiO_4$–$Ca_2Al_2SiO_7$ is therefore quaternary in nature, and the composition of the phases cannot be represented in the plane of the diagram but must be viewed in terms of the parent-tetrahedron.

The junction of the liquidus curves is

[24] Pp. 27–37 of ftn. 10.

[25] P. 31 of ftn. 10.

at 64 per cent $NaAlSiO_4$ at a temperature of 1,287° C. This pseudoeutectic is actually divariant, representing the trace of the boundary line between the fields of nepheline solid solution and melilite in the quaternary system. The junction of the carnegieite liquidus with the nepheline liquidus occurs at 67 per cent $NaAlSiO_4$ and at 1,297° C. Just as the pseudoeutectic is not invariant, so these liquidus curves are not univariant but trivariant, and the curves do not give the compositions of the liquids in equilibrium with the solid phases at the various temperatures.

THE BINARY SYSTEM $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$

The binary system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$ was first investigated by G. A. Rankin and F. E. Wright in their work on the system CaO–Al_2O_3–SiO_2.[26] The binary join has been revised by E. F. Osborn,[27] and his data are here used in constructing the equilibrium diagram of Figure 4. The system shows none of the complications of the preceding limiting systems. The eutectic is at 51.7 per cent anorthite and at a temperature of 1,387° ± 2° C.

THE THERMAL DATA

The thermal data necessary for establishing the equilibrium relationships in the system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$ are presented in Table 1, and the diagrams based on these results are shown in Figures 6 and 7. In Figure 6 are shown the isotherms at 50° intervals, while in Figure 7 the isotherms have been omitted, the stability fields have been labeled, and arrows have been inserted to indicate the directions of falling temperature. Figure 5 is a composition–refractive index[28] diagram of the homogeneous glasses prepared in this plane.

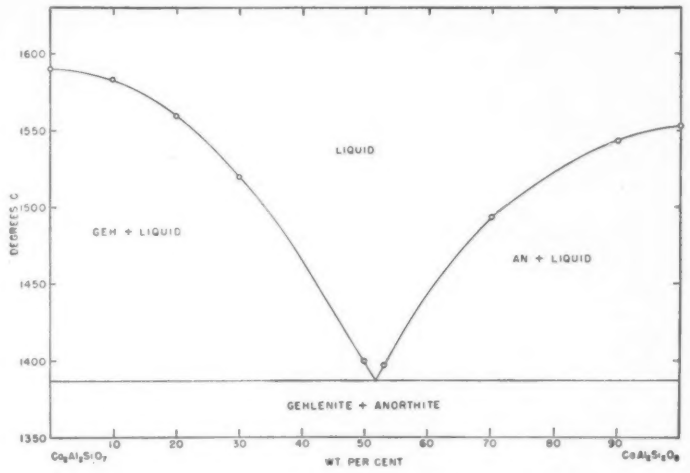

FIG. 4.—Equilibrium diagram for the binary system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$ (after Rankin and Wright, as revised by E. F. Osborn).

[26] "The Ternary System of CaO–Al_2O_3–SiO_2," *Amer. Jour. Sci.*, Vol. XXXIX (4th ser., 1915), pp. 1–79.

[27] Unpublished data.

[28] Sodium-vapor light was used for all refractive index measurements in this study.

NONTERNARY NATURE OF THE SYSTEM

The system is not a true ternary system; indeed, it is quaternary throughout, since a field of basic plagioclase is present rather than one of anorthite, a field of melilite instead of gehlenite, and carnegieite and nepheline also form solid solutions that cannot be expressed in terms of the three end-members. The field of β–Al_2O_3 encroaches upon the system as an extension of the quaternary field of alumina originating at the Al_2O_3 corner of the tetrahedron. The system is thus quite complex, the composition of any liquid upon crystallization does not remain in the plane defined by the three end-members; courses of crystallization can be discussed only in terms of the parent–soda-lime-alumina-silica system. A more complete discussion as to the nonternary behavior of mixtures in this plane will be presented in a following section on courses of crystallization.

THE LIQUIDUS RELATIONS

The five fields of the primary phases are, in order of decreasing size, melilite, plagioclase, nepheline solid solutions, carnegieite solid solutions, and β-alumina. The configuration of the fusion surfaces can be ascertained from the isotherms of Figure 6. The carnegieite surface dips rather sharply into the adjoining fusion surface of nepheline, which is relatively flat. The surfaces of melilite and plagioclase are convex and slope toward the center of the system. The fusion surface of β–Al_2O_3 is but tentatively indicated, owing to the somewhat uncertain data on the liquidus relations along the $NaAlSiO_4$–$CaAl_2Si_2O_8$ join.

A maximum is present on the boundary curve between the fields of melilite and nepheline solid solutions. This maximum, the position of which is indicated by the curvature of the 1,300° C. iso-

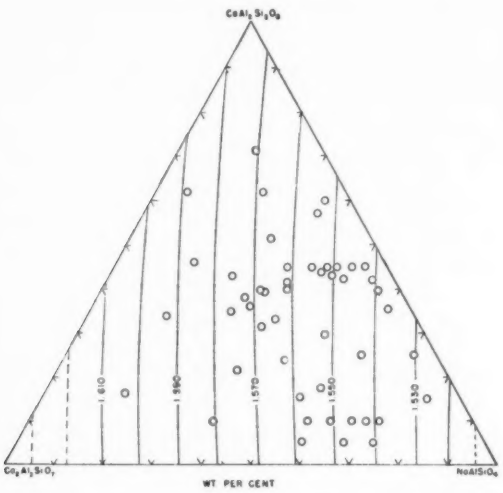

FIG. 5.—Composition–refractive-index diagram for the glass mixtures

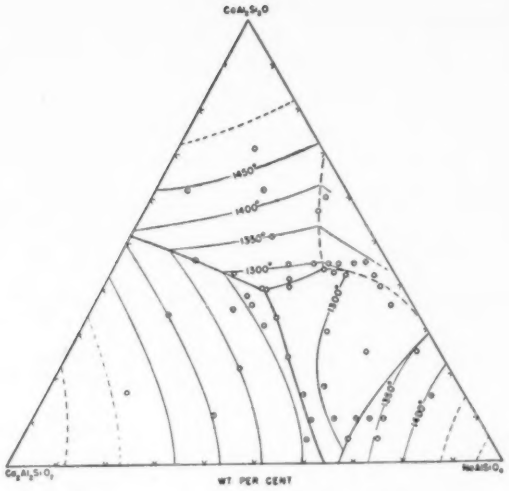

FIG. 6.—Equilibrium diagram for the system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$, with isotherms

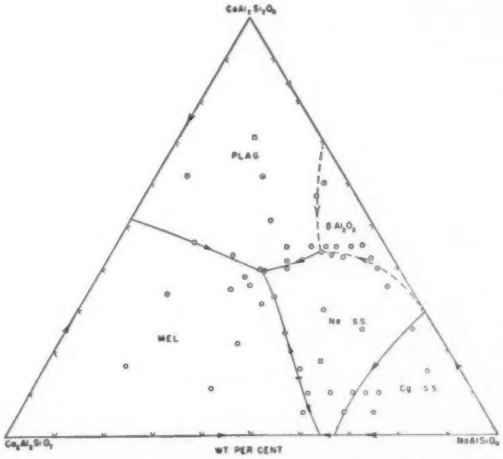

FIG. 7.—Equilibrium diagram for the system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$, with isotherms omitted

TABLE 1

THERMAL DATA FOR THE SYSTEM $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_3$

Weight Per Cent			Time (Minutes)	Temperature (° C.)	Final Condition
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$			
Primary-Phase Melilite					
31.3	32	36.7	30	1299	All glass
			30	1295	Glass, rare MEL
5.2	36.8	.58	30	1296	All glass
			30	1292	Glass, MEL
33.1	28.5	38.4	30	1278	All glass
			30	1274	Glass, NE, rare MEL
24	31	45	30	1288	All glass
			30	1284	Glass, NE, rare MEL
46	38.1	15.9	30	1328	All glass
			30	1324	Glass, MEL, rare AN
21.7	41.8	36.5	30	1352	All glass
			30	1348	Glass, MEL
11	52.6	36.4	30	1399	All glass
			30	1395	Glass, MEL
34	50	16	30	1407	All glass
			30	1403	Glass, rare MEL
36	32	32	30	1293	All glass
			30	1289	Glass, MEL
			30	1259	Glass, MEL, NE
			30	1242	Glass, MEL, NE, AN
38	32	30	30	1293	All glass
			30	1289	Glass, MEL
			30	1259	Glass, MEL, rare AN
			30	1255	Glass, MEL, AN, NE
35	$36\frac{1}{3}$	$28\frac{2}{3}$	30	1319	All glass
			30	1315	Glass, MEL
			30	1259	Glass, MEL
			30	1255	Glass, MEL, NE, AN
16.4	67.1	16.5	30	1491	All glass
			30	1486	Glass, MEL
Primary-Phase Anorthite					
71.3	13.4	15.3	30	1469	All glass
			30	1465	Glass and AN
39.7	28.1	32.2	30	1269	All glass
			30	1267	Glass, rare AN
			30	1264	Glass, AN, MEL
			30	1260	Glass, AN, MEL, NE

TABLE 1—*Continued*

Weight Per Cent			Time (Minutes)	Temperature (° C.)	Final Condition
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$			
Primary-Phase Anorthite—*Continued*					
45	20	35	30	1306	All glass
			30	1302	Glass, rare AN
			30	1259	Glass, AN, NE
			30	1251	Glass, AN, NE, MEL
45	15	40	30	1302	All glass
			30	1298	Glass, rare AN
			30	1288	Glass, AN
			30	1280	Glass, AN, NE
57	8	35	30	1376	All glass
			30	1372	Glass, rare β-Al_2O_3, AN
41.5	21.8	36.7	30	1283	All glass
			30	1279	Glass, rare AN
			30	1275	Glass, AN
			30	1271	Glass, AN, NE
			30	1250	Glass, AN, NE
			30	1242	Glass, AN, NE, MEL
46	38.1	15.9	30	1328	All glass
			30	1324	Glass, MEL, rare AN
43	32	25	30	1298	All glass
			30	1294	Glass, very rare AN
			30	1289	Glass, AN
			30	1285	Glass, AN, rare MEL
51.3	20.1	28.6	30	1352	All glass
			30	1348	Glass, AN
61.8	16.6	21.6	30	1422	All glass
			30	1416	Glass, AN
62	31.7	6.3	30	1449	All glass
			30	1445	Glass, AN
Primary-Phase Nepheline					
$15\frac{2}{3}$	32	$52\frac{2}{3}$	30	1295	All glass
			30	1291	Glass, NE
			30	1287	Glass, NE, MEL
33.1	28.5	38.4	30	1278	All glass
			30	1274	Glass, NE, rare MEL
24	31	45	30	1288	All glass
			30	1284	Glass, NE, rare MEL
42.4	9.9	47.7	30	1302	All glass
			30	1298	Glass, NE
43	12	45	30	1297	All glass
			30	1293	Glass, NE

TABLE 1—*Continued*

Weight Per Cent			Time (Minutes)	Temperature (° C.)	Final Condition
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$			
Primary-Phase Nepheline—*Continued*					
35.6	4.4	60	30	1329	All glass
			30	1325	Glass, NE
39.8	4.4	55.8	30	1319	All glass
			30	1315	Glass, NE
			30	1298	Glass, NE, rare β–Al_2O_3
5.2	28.4	66.4	30	1306	All glass
			30	1302	Glass, NE-CG
40	22.6	37.4	30	1273	All glass
			30	1269	Glass, NE
			30	1265	Glass, NE, AN
			30	1250	Glass, NE, AN
			30	1246	Some glass, NE, AN, MEL
17.4	27	55.6	30	1304	All glass
			30	1302	Glass, rare NE
29.7	20	50.3	30	1306	All glass
			30	1302	Glass, extremely rare NE
39⅓	27⅓	33⅓	30	1269	All glass
			30	1267	Glass, rare NE
			30	1263	Glass, NE, MEL
			30	1259	Glass, NE, MEL, rare AN
25.1	14.8	60.1	30	1327	All glass
			30	1323	Glass, NE
Primary-Phase Carnegieite					
5.1	22.6	72.3	30	1357	All glass
			30	1353	Glass, CG
25.1	4.4	70.5	30	1358	All glass
			30	1354	Glass, CG
5.2	28.4	66.4	30	1306	All glass
			30	1302	Glass, NE-CG
15.1	6.6	78.3	30	1406	All glass
			30	1402	Glass, abundant CG
Primary-Phase β–Al_2O_3					
45.2	4.4	50.4	30	1338	Glass, very rare β–Al_2O_3
			30	1342	All glass
45	7	48	30	1323	All glass
			30	1319	Glass, rare β–Al_2O_3
			30	1297	Glass, sparse β–Al_2O_3
			30	1280	Glass, NE, AN

TABLE 1—*Continued*

Weight Per Cent			Time (Minutes)	Temperature (° C.)	Final Condition
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$			
Primary-Phase β-Al_2O_3—*Continued*					
45	10	45	30	1302	Glass, rare β-Al_2O_3
			30	1306	All glass
45	12	43	30	1306	All glass
			30	1302	Glass, rare β-Al_2O_3
60	5	35	30	1393	All glass
			30	1389	Glass, rare β-Al_2O_3
			30	1384	Glass, β-Al_2O_3
			30	1376	Glass, β-Al_2O_3, AN
43.9	13.6	42.5	30	1294	All glass
			30	1292	Glass, very rare β-Al_2O_3
			30	1288	Glass, NE, AN, β-Al_2O_3
57	8	35	30	1376	All glass
			30	1372	Glass, rare β-Al_2O_3, AN
42.2	4.4	53.4	30	1322	All glass
			30	1318	Glass, rare β-Al_2O_3

YODER'S POINTS (UNPUBLISHED)

Weight Per Cent			Liquidus Temperature
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$	
Primary-Phase Nepheline			
10	33.3	56.7	1288° (checked), MEL at 1286°
10	28.8	61.2	1298° redetermined at 1302°
10	24.3	65.7	1314°
Primary-Phase Carnegieite			
10	21.6	68.4	1331°
10	18.9	71.1	1348°

therms in the two fields, is not very pronounced; it is but 3° higher than the temperature of the pseudoeutectic on the limiting $NaAlSiO_4$–$Ca_2Al_2SiO_7$ join. In a truly ternary system there is a maximum temperature on the boundary curve at the point where the line joining the composition of the two solid phases in equilibrium with a liquid intersects the boundary curve. This fact can sometimes be used as an indication of the composition of the solid phases, but no such interpretation can be made in the present case. The compositions of the phases do not lie in the plane of the system, and the solid solutions cannot be treated in ternary fashion. It does not necessarily follow that a composition on the gehlenite-nepheline side of the maximum will completely crystallize as some point "below" the maximum, i.e., in a direction away from anorthite, as it would if the maximum were present in the boundary curve of a ternary system.

The junction of the fields of plagioclase, melilite, and nepheline solid solutions is at a composition approximately $39\frac{1}{2}$ per cent anorthite, $27\frac{1}{2}$ per cent gehlenite, and 33 per cent nepheline and at a temperature of 1,266° C. This point is not a ternary eutectic, as crystallization is not completed at the temperature indicated (1,266°), nor does this point represent the composition of the last residual liquid. It is merely the intersection of the plane of the system with the univariant boundary curve between the quaternary fields of melilite, plagioclase, and nepheline solid solutions.

The boundary curves are not curves of univariant equilibrium but are divariant; the change in composition of a liquid in the system that is undergoing crystallization is not along a boundary curve, as it is in a ternary system. The nepheline solid solution–carnegieite solid solution boundary curve is not an isotherm, as the two extremities are not at the same temperature. Inversion from the high- to the low-temperature form begins at 1,352° C. at the $CaAl_2Si_2O_8$–$NaAlSiO_4$ join, and at 1,297° C. at the $Ca_2Al_2SiO_7$–$NaAlSiO_4$ join, with intermediate temperatures along the boundary between the two.

THE PLANE $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$ PLUS 10 PER CENT $CaSiO_3$

To obtain more definite information on courses of crystallization, and, by so doing, to obtain some information as to the composition of the mix-crystals (with particular reference to the melilites) that form from melts in this system, it was deemed desirable to trace the course of the boundary curve between the fields of plagioclase, melilite, and nepheline solid solutions within the quaternary system. As has been seen, the pseudoeutectic point marks the position and temperature of this curve as it passes through the plane of the system here studied.

The technique used was to locate the pseudoeutectic (the point where plagioclase, melilite, and nepheline solid solutions crystallize simultaneously) in some other plane of the tetrahedron. The plane of $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$ plus 10 per cent $CaSiO_3$ within the subsidiary tetrahedron anorthite-gehlenite-nepheline-wollastonite was chosen as a convenient one. Figure 8 represents the subsidiary tetrahedron, oriented as in Figure 1 but removed from the parent-tetrahedron and enlarged for clarity. The dotted lines indicate the plane of 10 per cent wollastonite. The location of the pseudoeutectic was determined in this plane with but three quenching runs, and the third composition that was made up fortunately corresponded exactly with the desired point on the

boundary curve. These data are given in Table 2.

There have now been located two points on the univariant boundary curve in the quaternary system between the fields of plagioclase, melilite, and nepheline solid solutions. The two four-phase points have the following parameters:

1. Composition = 39½ per cent anorthite, 27½ per cent gehlenite, 33 per cent nepheline; temperature = 1266° C.
2. Composition = 35.7 per cent anorthite, 17.8 per cent gehlenite, 36.5 per cent nepheline, 10 per cent wollastonite; temperature = 1248° C.

If this curve were to be accurately determined, numerous points within the quaternary system would have to be located. It is here assumed that a straight line joining the two determined points represents the boundary curve. If this curve is to be used as a reference line, the temperature gradient along it must also be known; and again the assumption is made that a linear relationship holds, defined by the temperatures of the two measured points. That these assumptions are not entirely correct can be shown by consideration of the system $CaSiO_3$–$CaAl_2Si_2O_8$–$NaAlSiO_4$.[29] This system, which forms one face of the subsidiary tetrahedron as shown in Figure 8, is encroached upon by the field of melilite. Therefore, at the junction of the fields of melilite, plagioclase, and nepheline solid solutions, there is located a third point on the quaternary boundary curve. Gummer's diagram[30] shows this point to be approximately 34 per cent anorthite, 38 per cent nepheline, and 28 per cent wollastonite, and at a temperature of 1,190° C. Consideration of the three points shows that the temperature drop along the boundary is not linear, and a projection of the compositions of the three indicates that the boundary is not straight. The departure from linearity between the values of 0–28 per cent $CaSiO_3$ is not considerable, however, and errors introduced by assuming linear values between 0 and 10 per cent $CaSiO_3$ are small enough to justify the assumptions. The complications introduced by solid solutions bring up the possibility of a temperature maximum along the boundary curve, in which case any assumptions as to temperature gradient are, of course, nullified. It is not probable that such a situation exists, however, in the limited extent of the boundary under consideration. Thus knowledge of the temperature at which nepheline solid solution and plagioclase simultaneously crystallize fixes the composition of the liquid at a point on the boundary curve corresponding to this temperature.

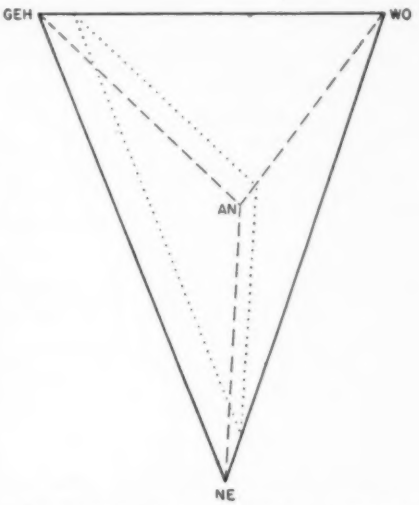

FIG. 8.—The subsidiary tetrahedron AN-GEH-NE-WO, showing (*dotted lines*) plane of 10 per cent WO.

COURSES OF CRYSTALLIZATION

The fact that the composition of the crystalline phases does not lie in the plane

[29] See ftn. 21.

[30] P. 515 of ftn. 21.

of the system and, consequently, that the residual liquids resulting from crystallization also leave this plane makes difficult the tracing of courses of crystallization. If the compositions of the solid solutions were precisely known, the courses of crystallization viewed in three dimensions within the quaternary system could be precisely described. The present state of uncertainty as to the composition of these solid solutions, with the possible exception of the plagioclase, makes this impossible.

The plagioclase-melilite-nepheline boundary curve was located for the purpose of obtaining data on the courses of crystallization and has been used in the present work to locate what may be called a "four-phase boundary" within the quaternary field of melilite. The technique of locating three-phase boundaries in a ternary system has been described by N. L. Bowen.[31] In the present investigation the method used was to locate a composition in the melilite field from which plagioclase and nepheline solid solutions simultaneously crystallized as secondary phases. The significance of the simultaneous crystallization of the secondary phases is that the crystallization of the primary phase, melilite, results in a liquid of changing composition (increasingly deficient in the melilite composition), the composition path of which directly hits the *melilite-plagioclase-nepheline boundary curve* in the quaternary system. Thus the location of such a point in the field of melilite, coupled with the observation of the temperature at which the plagioclase and nepheline solid solution crystal-

TABLE 2

THERMAL DATA FOR THE PLANE $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$ + 10 PER CENT $CaSiO_3$

WEIGHT PER CENT				TIME (MINUTES)	TEMPERATURE (° C.)	FINAL CONDITION
$CaAl_2Si_2O_8$	$Ca_2Al_2SiO_7$	$NaAlSiO_4$	$CaSiO_3$			
Primary-Phase Melilite						
$37\frac{2}{3}$	$21\frac{2}{3}$	$30\frac{2}{3}$	10	30	1275	All glass
				30	1271	Glass, MEL
				30	1263	Glass, MEL, AN
Primary-Phase Anorthite						
37.2	19.7	33.1	10	30	1267	All glass
				30	1263	Glass, AN, very rare MEL
Simultaneous Phases Anorthite-Melilite-Nepheline						
35.7	17.8	36.5	10	30	1250	All glass
				30	1246	Glass, MEL, NE, sparse AN

[31] "The Crystallization of Haplobasaltic, Haplodioritic, and Related Magmas," *Amer. Jour. Sci.*, Vol. XL (4th ser., 1915), pp. 171–74.

lize, fixes two points on a crystallization path in the melilite field. As described above, the boundary curve has been located within the quaternary system, and the temperature of appearance[32] of the secondary phases locates the point on the boundary curve that is "hit" by the crystallizing liquid. A line joining these two points must pass through the composition of the crystals that have separated from the liquid. The above considerations are applicable in principle to the field of nepheline, although determinations in this field have not been carried out.

Examination of Figure 6 shows three points near the pseudoeutectic in the field of melilite. These points, along with others to be subsequently discussed, are reproduced in Figure 9, which is an enlarged section of the equilibrium diagram.

If the system were ternary, the final temperature of consolidation of these three compositions would be 1,266°, and at the composition of the eutectic. Runs made on point 1 (Fig. 9) did not show nepheline until approximately 1,260° C., and plagioclase appeared at approximately 1,245° C. In point 2, plagioclase was present at approximately 1,260° C., and nepheline appeared at 1,255° C. Point 3 represents a composition in which plagioclase and nepheline solid solutions appear simultaneously and at a temperature of 1,257° C. Thus the quaternary boundary curve is encountered at a temperature of 1,257° C. for this particular initial composition. It has been shown that this boundary curve passes through the plane of 10 per cent wollastonite at 1,248° C. Assuming a linear temperature relationship, 1,257° C. is halfway between the two reference temperatures of 1,248° and 1,266° C.; and thus the boundary curve is encountered at approximately the plane of 5 per cent wollastonite within the subsidiary tetrahedron *AN-GEH-NE-WO*. This point will be more fully discussed in the following section, but it can be seen from examination of Figure 1 that this course of crystallization is roughly toward the SiO_2 corner of the main tetrahedron.

If the nepheline solid solutions were ternary, point *4* of Figure 9 would also consolidate at 1,266° C. Plagioclase appears in runs on this point at 1,267° C., and melilite does not appear until the temperature has fallen to 1,248° C. Point *5*, in the plagioclase field, precipitates nepheline at 1,273° C. and melilite at approximately 1,246° C., whereas in point *6* melilite appears at approximately 1,255° C. The fact that melilite does not appear in runs made on points *5* and *6* until temperatures well below that of the pseudoeutectic are reached is indicative of the rather strongly nonternary nature of the nepheline solid solutions. Even in compositions close to the pseudoeutectic, there is a considerable difference in tem-

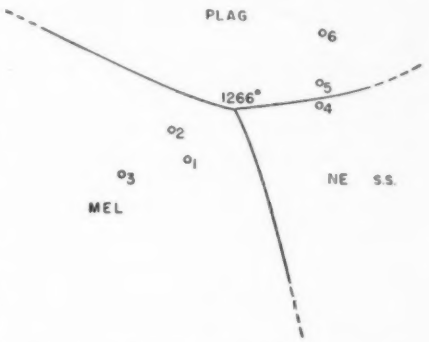

FIG. 9.—Enlarged portion of the equilibrium diagram for the system $CaAl_2Si_2O_8$–$Ca_2Al_2SiO_7$–$NaAlSiO_4$.

[32] If devitrified mixtures are used, this temperature is that of the simultaneous disappearance of the secondary phases.

perature between that of the pseudo-eutectic and that of appearance of the third crystalline phase, indicating that the composition of the liquid is well out of the plane of the system.

No attempt has been made to follow courses of crystallization within the fields of β–Al_2O_3 or carnegieite. In general, compositions within the field of β–Al_2O_3 will follow a path away from the Al_2O_3 corner of the tetrahedron.

THE SUBSTANCES IN SOLID SOLUTION

With the exception of quartz, all rock-forming minerals are members of solid-solution series. Pure end-members of solid-solution series are rarely, if ever, found in nature; and it has been amply demonstrated that the pure end-members are never formed in synthetic melts when there are substances present with which they may form solid solutions. The nature of the substances in solid solution in the nepheline and carnegieite of this system is unsettled, but certain data here found tend to limit the possibilities at least with respect to nepheline. The "four-phase boundary" in the melilite field has, moreover, brought to light the probable existence of a hitherto unknown "molecule" in solid solution with gehlenite.

ANORTHITE

It was earlier thought that relatively pure anorthite could form in melts containing nepheline, but it is now known that the pure end-members of the plagioclase series cannot form in melts containing both soda and lime. Although few optical data could be obtained on the feldspars because of their small size and platy habit, enough were obtained to indicate that an anorthite-rich plagioclase rather than pure anorthite is formed. Crystals formed in a mix with but little soda (approximately 6 per cent nepheline, 62 per cent anorthite, 32 per cent gehlenite) had a maximum index that very closely matched the glass at a value of $n = 1.588$, indicating that virtually pure anorthite is here formed. At a somewhat higher soda content (16 per cent nepheline, 46 per cent anorthite, 38 per cent gehlenite), the maximum plagioclase index again matched the index of the glass, at a value of 1.586. In two mixes near the plagioclase-nepheline boundary curve maximum values of $n = 1.585$ were observed, the nepheline content here reaching 37 per cent. The value of 1.585 for the lowest observed maximum plagioclase index indicates that the anorthite content probably does not drop below 95–96 per cent in this system.

Extinction angles of 72° and 74° on two well-formed crystals were observed, as measured from the albite twinning lamellae to *a*. These crystals were those formed from melts with the 37 per cent nepheline content. Reference to standard tables indicates an anorthite content of approximately 98 per cent, as against 95–96 per cent on the basis of index of refraction. No discrepancy exists, however, as recent work has shown that plagioclases formed at high temperatures do not have the same composition–extinction-angle relation as do the low-temperature plagioclases, for which standard tables have been compiled.[33, 34] Plagioclases formed at high temperatures show an apparently greater anorthite content on the basis of extinction angle, as is here evident.

[33] A. Köhler, "Die Abhängigkeit der Plagioklasoptik vom vergangenen Warmeverhalten," *Min. u. petrog. Mitt.*, Vol. LIII (1942), pp. 159–79.

[34] See bibliography in paper by Christoffer Oftedahl, "High Temperature Optics in Plagioclases of the Oslo Region," *Norsk. geol. Tids.*, Vol. XXIV (1944), pp. 75–78.

β-ALUMINA

The probable composition of β–Al_2O_3 has been discussed.

NEPHELINE

Several substances are known to enter into solid solution with nepheline in considerable amount. A complete series of solid solutions between $KalSiO_4$ and $NaAlSiO_4$ exists,[35] but potash compounds are excluded in this study. Experimental work has shown that 35 per cent each of albite[36] and anorthite[37] are taken into solid solution. The plagioclases are sufficient to account for the excess silica, as well as the calcium that is found in natural nephelines; but A. N. Winchell[38] has suggested that the molecule $CaAlAlO_4$ may also play a part in the composition.

The high indices found by Smalley in his work on the system $NaAlSiO_4$–$Ca_2Al_2SiO_7$[39] are considerably above those brought about by solid solution of anorthite in nepheline. The inversion relations of nepheline and carnegieite were also such that anorthite could not be considered as the material entirely responsible. In the present study no negative nephelines were observed; those formed from melts closest to the field of carnegieite (at approximately 60 per cent nepheline) were sensibly isotropic, those precipitated from all other melts poorer in the nepheline molecule were positive, becoming more strongly birefringent as the $NaAlSiO_4$ content of the liquid decreased. The nepheline indices, as in Smalley's system, increase with increasing $Ca_2Al_2SiO_7$ content of the melt. Isotropic crystals with $n = 1.540$ were observed in the mixture at 5 per cent $Ca_2Al_2SiO_7$, 60 per cent $NaAlSiO_4$, 35 per cent $CaAl_2Si_3O_8$; and values of $\omega = 1.548$, $\epsilon > 1.548$, were observed in crystals near the plagioclase-nepheline boundary curve at a value of roughly 20 per cent $Ca_2Al_2SiO_7$.

It was noted that, in mixtures from which nepheline crystallized as a primary or secondary phase and melilite as the tertiary phase, the temperature of appearance of the melilite was considerably below that of the pseudoeutectic. The interpretation was that the quaternary boundary curve between the fields of melilite, plagioclase, and nepheline solid solutions was reached in the direction of falling temperature. That is, the temperature along this boundary falls in the direction of the direction of the anorthite-nepheline-wollastonite face of the subsidiary tetrahedron, and the composition of the liquid must change in this direction upon crystallization of nepheline solid solutions. Thus plagioclase cannot be the sole substance in solid solution in the nephelines of this system. In addition, anorthite or any substance near anorthite in composition would not account for the strong nonternary tendency that is shown in the nepheline solid solutions; the plagioclase compositions lie nearly in the same plane as that of this system; anorthite, of course, lies in the plane.

It thus appears that at least part of the material entering into solid solution with nepheline lies on that side of the system that is toward the Al_2O_3 corner of the main tetrahedron. Sodium or calcium aluminates would fulfil this requirement. The $CaAlAlO_4$ molecule proposed by Winchell might well be a substance en-

[35] N. L. Bowen, "The Sodium-Potassium Nephelites," *Amer. Jour. Sci.*, Vol. XLIII (4th ser., 1917), pp. 115–32.

[36] P. 106 of ftn. 7.

[37] P. 563 of ftn. 4.

[38] *Elements of Optical Mineralogy*, Part II (3d ed.; New York: John Wiley & Sons, Inc., 1933), p. 297.

[39] See ftn. 10.

tering into solid solution with nepheline in this system. It is also known that $Na_2Al_2O_4$ forms a solid-solution series with carnegieite,[40] and it is not unlikely that it can also form solid solutions with nepheline. The establishment of "four-phase boundaries" in the field of nepheline might shed more light on the exact nature of the solid solutions.

At first sight it appears peculiar that no evidence of $CaAlAlO_4$ and/or $Na_2Al_2O_4$ is found in naturally occurring nephelines. An excess of SiO_2 is usually present, and the above compounds would make for undersilicated nepheline. The system studied is deficient in silica, however, as compared to natural magmas, and it is likely that in magmas the silica content is always sufficient so that feldspar forms solid solutions with nepheline rather than the aluminates. It is more probable, however, that at the high temperatures of this work, probably higher than those of magmas, solid solutions form that are not found in nature.

CARNEGIEITE

This investigation has produced no new evidence as to the nature of the carnegieite solid solutions. Smalley[41] has observed a marked index rise and inversion lowering in the system $NaAlSiO_4$–$Ca_2Al_2SiO_7$. Inversion lowering is very unusual in silicates, as it requires a greater amount of solid solution in the high-temperature form than in the low-temperature form of an enantiotropic pair. However, as the carnegieite solid solutions undoubtedly present a problem very similar to that of the nepheline mixcrystals, the situation is perhaps not too obscure. Here anorthite is unquestionably out of consideration as the responsible agent, as it is taken up by carnegieite only to the extent of 5 per cent, whereas nepheline will take 35 per cent.[42] C. E. Tilley[43] found that Na_2SiO_3 entered into carnegieite solid solutions to the extent of 24 per cent but not at all into nepheline. The changes in birefringence and index rise were not consistent with those found by Smalley, however, and it is unlikely that Na_2SiO_3 plays a part in this system. Smalley found that the trend of the solid solutions was to increase in concentration with increase in the $Ca_2Al_2SiO_7$ content of the mixture and suggested that the sodium aluminate ($Na_2Al_2O_4$) was responsible; the decrease in silica brought about by the addition of gehlenite would account for the trend rather than the addition of lime, with formation of a lime-bearing solid solution. This view is in line with the discussion of the nepheline solid solutions, and, in addition, it may be that the $CaAlAlO_4$ molecule is also present in the mixcrystals.

GEHLENITE

The indices of the melilites that crystallize in this system are significantly lower than those of pure gehlenite. Gehlenite has indices of $\omega = 1.669$ and $\epsilon = 1.658$. The melilite formed from a mixture of composition anorthite 16 per cent, gehlenite 67 per cent, and nepheline 17 per cent had indices of $\omega = 1.665$, $\epsilon' = 1.656$; and in a composition of anorthite 38 per cent, gehlenite 32 per cent, and nepheline 30 per cent the indices were $\omega = 1.656$, $\epsilon' = 1.645$; at anorthite 5 per cent, gehlenite 37 per cent, and nepheline 58 per cent, $\omega = 1.651$, $\epsilon' = 1.641$. A progressive decrease

[40] Unpublished work of J. F. Schairer and N. L. Bowen.

[41] See ftn. 10.

[42] P. 563 of ftn. 4.

[43] "The Ternary System Na_2SiO_3–$Na_2Si_2O_5$–$NaAlSiO_4$" *Min. u. petrog. Mitt.*, Vol. XLIII (1933), p. 414.

in index with increasing soda content is thus observed, a decrease that was also found by Smalley to exist in the limiting $Ca_2Al_2SiO_7$–$NaAlSiO_4$ system. No decrease in birefringence was observed.

The natural melilites have complex solid-solution relations, and this complexity has elicited considerable discussion. A complete series of solid solutions exists between $Ca_2Al_2SiO_7$ and $Ca_2MgSi_2O_7$ (akermanite); but for purposes of this discussion the magnesia-bearing varieties, as well as the iron melilites, will be neglected. In 1924, A. N. Winchell[44] took up the melilite problem from the viewpoint of volume isomorphism and largely discredited the sarcolite molecules which had been proposed earlier by W. T. Schaller[45] and partially verified by A. F. Buddington.[46] Winchell pointed out the essential R_5O_7 character of the melilites and proposed the molecules $Na_2Si_3O_7$ and $Ca_3Si_2O_7$ as theoretically capable of entering the gehlenite structure. In addition, the possibility of the presence of several sodium-aluminum silicates and a calcium-aluminum silicate was considered. Berman[47] in 1929 reviewed the problem and modified Winchell's fundamental R_5O_7 to $X_2Y_3O_7$. He also changed Winchell's $Ca_3Si_2O_7$ to $CaSi_3O_7$. The melilite molecules, as seen by Berman (omitting akermanite) are shown in the accompanying tabulation.

Gehlenite	$Ca_2Al_2SiO_7$
Soda melilite	$Na_2Si_2O_7$
Submelilite	$CaSi_3O_7$

[44] "The Composition of Mellilite," *Amer. Jour. Sci.*, Vol. VIII (5th ser., 1922), pp. 375–87.

[45] "Mineralogic Notes," Ser. 3, *Bull. U.S. Geol. Surv., No. 610* (1916), pp. 109–28.

[46] "On Some Natural and Synthetic Melilites," *Amer. Jour. Sci.*, Vol. III (January, 1922), pp. 35–87.

[47] Harry Berman, "Composition of the Melilite Group," *Amer. Min.*, Vol. XIV, No. 11 (November, 1929), pp. 389–407.

On the basis of chemical analyses Berman stated that the "soda melilite" does not exceed 25 per cent in natural occurrences and the "submelilite" not over 10 per cent.

The location of the quaternary boundary curve between the fields of melilite, plagioclase, and nepheline solid solutions and the determination of the "four-phase boundary" discussed in the section on courses of crystallization was made principally to obtain information on the nature of the melilite mix-crystals in this system.

The essential data with respect to the "four-phase boundary" are here summarized. A liquid represented by the composition of point *3* in Figure 9, upon crystallizing melilite, hits the quaternary boundary curve at 1,257° C., as evidenced by simultaneous crystallization of plagioclase and nepheline as secondary phases. Two points are thus located; the initial composition (point *3*, Fig. 9), and that point on the boundary curve representing the composition of the liquid at the time when sufficient melilite has crystallized to attain this composition. A straight line, drawn from this point on the boundary curve through the initial composition, points to the composition of the melilite crystallizing at the time that the boundary curve is reached. The composition of the initial point is known, and that of the point on the boundary curve can be located by making the assumptions of a straight boundary curve with linear temperature drop, as earlier discussed. As the temperature is 1,257°, half of the difference between the planes of 0 and 10 per cent wollastonite can be assumed to be in the plane of 5 per cent wollastonite. Calculation of this point gives the following oxide percentages,

and listed with it is the initial composition of the mixture:

		Per Cent
Initial composition	CaO	21.92
	Na_2O	6.26
	Al_2O_3	36.62
	SiO_2	35.20
Point on boundary curve	CaO	19.11
	Na_2O	7.67
	Al_2O_3	34.64
	SiO_2	38.55

Figure 10 represents the subsidiary tetrahedron, as in Figure 8. The quaternary boundary curve is diagrammatically indicated by the slightly curved line. Points *a* and *b* represent, respectively, the initial composition (point 3, Fig. 9) and the point on the boundary curve hit by this point upon crystallization. The arrow, joining the two, points to the composition of the melilite forming when point *b* is reached.

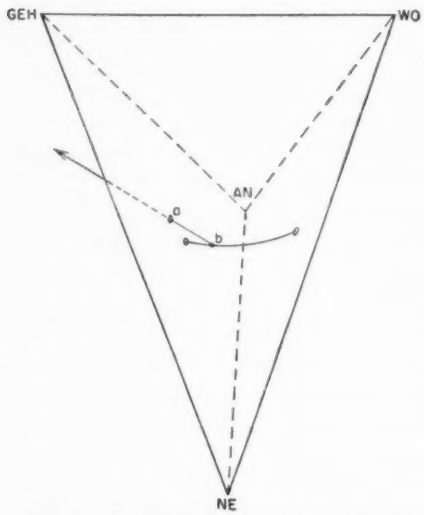

FIG. 10.—The subsidiary tetrahedron AN-GEH-NE-WO, showing quaternary boundary curve and four-phase boundary.

It is evident that the melilite solid solution crystallizing from the composition of point *a* is represented by some point "below" the plane of this system, i.e., on the side toward the Al_2O_3 corner of the main tetrahedron. Thus none of the hypothetical molecules proposed by Winchell or Berman enter into solid solution in this system. Considering pure gehlenite, $Ca_2Al_2SiO_7$, the following replacements can be made:

$CaCaAlAlSiO_7$ (gehlenite)
$CaAlAlAlAlO_7$ (Al for Ca and Al for Si)

This molecule, conforming to the gehlenite structure, can be written $CaO \cdot 2Al_2O_3$ and will be referred to as "1:2." The 1:2 "molecule" is not known to exist as an independent compound,[48] but this fact does not preclude its entering into solid solution as a component. If it does form a solid solution with gehlenite, the composition of the mixcrystals would be represented by a line joining the two components within the principal tetrahedron. The extension of the join between points *a* and *b* (Fig. 10) should then meet this line at some point representing the composition of the mixcrystals formed from a melt of composition *a*.

An orthographic projection of the lines representing the joins between *a* and *b* and gehlenite–1:2 shows that they come very close to intersecting.[49] This fact could also be proved by analytical means, either by obtaining the equations of the two lines or by showing that all four points lie in a plane.

[48] P. 306 of ftn. 2.

[49] A precise join would be fortuitous, because of experimental errors, here listed:

1. The primary phase of point 3, Fig. 9, is melilite, the high relief of which tends to mask the appearance of the secondary phases. The determination of simultaneous phases, as well as the precise temperature of appearance, is difficult.

2. Points *a* and *b* are relatively close together in terms of the main tetrahedron; thus a slight error in the join between them is magnified in the extension of the join. If a point such as *a* were located at a greater distance from the boundary curve, the

On the basis of this single determination, it thus appears that the 1:2 compound of lime-alumina is the substance in solid solution in gehlenite crystallizing from this system. Moreover, if any other hypothetical "molecule" enters the mix-crystals, it must do so in very small amounts.

As was postulated for the nepheline solid solution, it is again a subsilicious phase that precipitates from melts of the present investigation. No such silica-poor melilites are found in nature. Natural magmas are not comparable in composition to melts in this system, however; and, as previously discussed, the temperatures of crystallization are considerably lower in magmas. This difference of composition of the natural magmas presumably results in the formation of solid solutions of a nature other than those containing the 1:2 molecule, and the understanding of the complexity of the melilites is by no means completely resolved by this investigation.[50]

identification of secondary phases (1, above) would be more difficult.

3. The assumption of a straight boundary curve with linear temperature drop is not strictly true.

4. There are experimental errors of temperature measurement and composition synthesis.

[50] Although the results of this investigation as interpreted above indicate solid solution of $CaO \cdot 2Al_2O_3$ in gehlenite, certain difficulties are encountered in this supposition. No solid solutions of this nature were encountered in the investigation of the system CaO–Al_2O_3–SiO_2 (ftn. 26). The proposed substitution of Al for Ca in the solid solutions is rather doubtful, owing to the small size of the Al atom. It is therefore important to note that another interpretation is possible: *if* a maximum temperature is present on the melilite-nepheline-plagioclase boundary curve, the melilites crystalizing from this system may be *enriched* in Si and Na. Thus, if a temperature of 1,257° exists on the low-silica side of the plane of the system, the "molecule" in solid solution in gehlenite could be the $Na_2Si_3O_7$ proposed by Berman, which, it must be admitted, is more likely from a structural point of view. It is hoped that this point can be cleared up in a future investigation.

PETROLOGIC SIGNIFICANCE

Complete knowledge of the equilibrium relations within the soda-lime-alumina-silica system is of great importance in the understanding of the problems of igneous petrology, and the present paper is a contribution toward that end. These four oxides constitute, by weight, 90 per cent of the average of 546 granites, 85 per cent of the average of 43 nepheline syenites, and 77 per cent of the average of 161 basalts.[51] The minerals—quartz, plagioclase feldspars, nepheline, wollastonite, gehlenite, corundum, sillimanite, mullite, grossularite, larnite, and jadeite—are formed of these four oxides, although the last five are formed only under "metamorphic conditions." Knowledge of the four oxide systems is also of prime importance in certain technological fields.

The equilibrium relations as determined in this study contribute to the knowledge of the relations within the main system. All seven faces of the subsidiary double tetrahedron have now been investigated.[52, 53] It has been mentioned that the melts within the plane here studied do not correspond in composition to any natural magmas. The closest approach is to be found in magmas that formed certain basic alkaline rocks, such as the tephrites, melilite-nephelinites, urtites and ijolites, turjaites, and melilite basalts. All these rocks contain greater or lesser amounts of a pyroxene and numerous accessory

[51] R. A. Daly, *Igneous Rocks and the Depths of the Earth* (New York: McGraw-Hill Book Co., Inc., 1933, pp. 9–28.

[52] See bibliography in paper by N. L. Bowen, "Phase Equilibria Bearing on the Origin and Differentiation of Alkaline Rocks," *Amer. Jour. Sci.*, Vol. CCXLIII*A* (Daly Vol.) (1945), p. 83.

[53] The anorthite-gehlenite-wollastonite face is based on unpublished work of V. C. Juan.

minerals. The melilite of this system is also less complex than the natural mineral, because of the absence of magnesia and iron.

The strong tendency of gehlenite, nepheline, and anorthite to form solid solutions is again demonstrated in the present study; and, because of the nonternary nature of the solid solutions, the system must be considered in terms of the four constituent oxides. Crystallization of the nepheline and melilite solid solutions results in a liquid enriched in SiO_2 and Na_2O; and, once the quaternary boundary curve between the fields of melilite, plagioclase, and nepheline is reached, fractional crystallization could carry the residual liquid toward the plane of anorthite-nepheline-wollastonite, which is common to the subsidiary double tetrahedron. The fact that the field of melilite plunges through this boundary face, as discussed by Bowen,[54] results in a quaternary reaction point within the adjoining tetrahedron, anorthite-wollastonite-nepheline-albite, and permits fractionation to continue so that residual liquids within this portion of the main tetrahedron can result. Bowen[55] points out that fractionation can continue until the liquid reaches a composition within that portion of the quaternary system outlined by the compounds nepheline-albite-devitrite ($Na_2O \cdot 3CaO \cdot 6SiO_6$)–sodium disilicate, the crystalline assemblages running the gamut from simplified melilite-nephelinites through simplified tephrites and phonolites to alkali rhyolites, with a soda-rich liquor remaining.

The encroachment of a field of alumina upon the anorthite-nepheline join is suggestive in connection with the occurrence of corundum in certain anorthosites and nepheline-bearing syenites.

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[54] See reference cited in ftn. 52.

[55] See reference cited in ftn. 52.

THE COMPOSITION OF METEORITIC MATTER

I. THE COMPOSITION OF THE SILICATE PHASE OF STONY METEORITES

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ABSTRACT

A statistical study has been made of the analyses of the silicate phases of 107 selected stony meteorites. Frequency-distribution curves have been plotted, and arithmetic means, together with standard deviations and precisions, have been calculated for the major constituents.

INTRODUCTION

At the present time, meteorites constitute one of the more important sources of information concerning the composition of the solar system. A thorough study of the composition of meteoritic matter can, conceivably, give valuable clues for the solution of such diverse problems as the geochemical distribution of the elements, the process of planet formation, and the origin of the elements.

In 1938, V. M. Goldschmidt,[1] utilizing primarily chemical data obtained on meteorites, compiled a table of "cosmic abundances" of nuclear species, which is of particular interest to nuclear physicists and astrophysicists because of its bearing upon the problems of the origin of the elements and the relationships which may exist between "cosmic abundance" and nuclear properties. Since that time, interest in the problem of the origin of the elements has progressed to a point at which further inquiry into the experimental and theoretical foundations for such a compilation is desirable. This paper represents the first phase of such an inquiry.

As our first step, we have attempted to answer two questions: (1) How accurately can the average composition of the silicate phase of stony meteorites be expressed at the present time? (2) For a given constituent, how many meteorites must be analyzed if the probable error of the arithmetic mean of the concentration is to lie below a certain fixed value? We have attempted to answer these questions by analyzing statistically the available data on the major constituents of stony meteorites.

THE PRESENT INQUIRY

In 1916, G. P. Merrill[2] averaged the composition of 59 stone meteorites, most of them known falls. Goldschmidt,[3] in his compilation of relative abundances, used Merrill's averages, after first subtracting the iron-nickel and sulphide phases, in that manner obtaining the average composition of the silicate phase. Since Merrill's averages were obtained, a sufficient number of additional meteorite falls have been analyzed to permit a statistical study of the distribution of the elements in the silicate phase.

The individual analyses for 107 selected stony meteorites are given in Table 1. Of the analyses, 58 are from

[1] "Geochemische Verteilungsgesetze der Elemente. IX. Die Mengenverhältnisse der Elemente und der Atom-Arten," *Skrifter utgitt av Det Norske Videnskaps-Akademi i Oslo. I Mat.-Naturv. Klasse, 1937*, No. 4 (1938).

[2] "Report on Researches on the Chemical and Mineralogical Composition of Meteorites, with Especial Reference to Their Minor Constituents," *Mem. Nat. Acad. Sci.*, Vol. XIV (1916).

[3] See ftn. 1.

TABLE 1

TABLE OF SELECTED ANALYSES OF STONY METEORITES

METEORITE*	SILICATE PHASE															IRON-NICKEL PHASE					REF.
	SiO_2	MgO	Fe†	Al_2O_3	CaO	Na_2O	K_2O	Cr_2O_3	NiO	MnO	CoO	TiO_2	H_2O	P‡	O§	Total	Fe	Ni	Co	S	Total	
Aguila Blanca, Argentina	48.19	27.79	9.86	3.95	2.11	1.71	0.26	0.75	2.34	0.22	Tr.	Tr.	.....	0.25	2.88	100.39	15.58	1.98	0.03	1.06	18.65	1
Alfianello, Italy	41.58	25.89	21.15	1.97	0.98	1.20	0.27	0.46	.....	0.14	.....	.....	.....	0.17	6.06	99.87	5.74	1.14	0.08	2.54	9.50	2
Allegan, Mich.	48.57	30.56	9.15	3.54	2.40	0.92	0.32	0.74	Tr.	0.25	.....	0.11	0.35	0.29	2.71	99.91	23.31	1.81	0.15	1.84	28.11	2
Angra dos Reis, Brazil	44.51	10.18	6.74	8.84	24.83	0.26	0.19	Tr.	.....	.....	.....	2.42	.....	0.10	1.96	100.03	0.81	.....	.....	0.45	1.26	2
Baroti, India	47.06	29.31	13.25	2.85	2.12	1.08	0.05	0.21	.....	Tr.	.....	0.19	0.20	0.23	3.87	100.42	12.45	0.75	0.03	2.47	15.70	2
Beaver Creek, Canada	48.28	30.61	10.75	2.80	2.27	1.02	0.13	0.27	0.04	0.30	.....	0.09	0.36	0.24	3.16	100.33	18.74	1.50	0.09	1.84	22.17	2
Bettrechies, France	47.71	27.96	16.79	0.22	2.56	0.64	0.22	.....	0.25	0.15	0.02	0.02	.....	0.03	4.95	101.52	14.64	0.71	0.07	3.87	19.29	3
Bishopville, S.C.	57.32	33.68	1.22	1.72	2.03	1.04	0.09	.....	0.54	0.19	Tr.	.....	2.00	.....	0.35	100.18	0.18	0.30	.....	0.04	0.52	2
Bjurbole, Finland	46.97	29.46	12.27	2.92	2.08	1.42	0.37	0.67	0.08	0.14	.....	.....	.....	0.16	3.52	100.06	9.84	0.72	0.04	1.98	12.58	2
Bluff, Tex.	39.89	27.44	19.59	2.30	2.33	.....	.....	.....	1.68	0.48	0.17	.....	.....	0.20	5.67	99.75	3.47	0.65	0.09	1.30	5.51	2
Bowden, S. Africa	47.55	30.10	10.25	3.77	2.89	0.99	0.14	0.42	.....	0.31	.....	0.16	0.35	.....	3.10	100.03	19.86	1.49	0.06	2.15	23.56	4
Cape Girardeau, Mo.	46.53	31.06	11.45	2.97	1.81	1.12	0.14	c 60	.....	.....	.....	.....	0.61	0.29	3.37	99.95	20.07	1.32	0.11	2.07	23.57	2
Colby, Wis.	45.28	32.16	12.81	3.10	—	1.21	0.15	0.55	0.23	0.49	0.02	.....	.....	0.20	3.74	99.94	5.73	0.07	0.02	2.76	8.58	5
Collescipoli, Italy	42.51	26.02	15.16	7.90	2.20	1.79	0.32	.....	.....	.....	.....	.....	.....	.....	4.34	100.24	17.04	1.49	0.09	.....	18.62	2
Cranstad, S. Africa	48.78	30.93	9.34	3.16	1.90	0.88	0.09	0.48	0.13	0.05	.....	0.39	0.45	0.25	2.75	99.58	20.00	1.55	0.23	1.88	23.66	6
Crumlin, Ireland	47.49	29.57	12.00	2.49	2.38	1.25	0.13	0.70	.....	0.31	.....	0.20	.....	0.23	3.51	100.26	11.09	1.15	0.08	1.91	14.23	7
Cullison, Kan.	47.27	31.64	9.42	5.68	0.83	2.41	0.23	0.05	.....	0.39	.....	.....	.....	0.01	2.70	100.63	21.27	1.80	0.10	2.18	25.35	2
Daniels Kuil, S. Africa	58.47	32.88	0.27	2.71	2.50	0.97	0.23	0.69	0.17	0.03	.....	0.18	0.52	Tr.	0.08	99.80	28.85	1.78	0.12	3.46	34.21	6
Ekerby, Sweden	44.29	28.72	14.78	2.27	2.49	1.22	0.26	0.58	.....	0.31	.....	0.28	0.04	0.28	4.34	99.86	16.93	1.73	.....	1.90	20.56	8
Elm Creek, Kan.	36.75	25.66	20.30	3.10	1.62	—	—	0.35	.....	0.01	.....	.....	5.10	.....	5.81	98.70	5.94	0.77	0.09	.....	6.80	2
Estacado, Tex.	43.56	27.65	14.68	4.38	3.64	2.52	0.39	—	.....	—	.....	—	.....	0.18	4.20	101.20	14.68	1.60	0.08	1.37	17.73	2
Farmington, Kan.	45.74	29.95	14.04	2.05	2.00	0.84	0.13	0.66	0.37	0.18	.....	.....	.....	.....	4.02	99.98	9.86	0.94	0.06	1.82	12.68	2
Felix, Ala.	36.39	21.40	22.09	3.51	5.91	0.67	0.15	0.87	1.09	0.74	.....	.....	0.17	.....	6.33	99.32	5.62	0.36	0.08	1.73	7.79	2
Fisher, Minn.	43.69	29.38	14.40	4.79	2.19	Tr.	—	0.54	0.23	0.38	.....	.....	.....	.....	4.13	99.73	9.72	1.61	0.08	Tr.	11.41	2
Florence, Tex.	46.69	30.22	10.94	3.57	3.59	0.50	0.74	0.23	0.18	0.70	.....	Tr.	.....	0.26	3.21	100.83	19.29	1.49	0.07	1.84	22.69	9
Forest City, Iowa	47.87	32.18	10.71	2.80	1.90	1.09	0.08	0.13	0.19	.....	.....	.....	.....	.....	3.07	100.02	22.02	1.19	0.13	2.25	25.59	2
Forksville, Va.	41.30	28.40	18.80	3.25	1.46	0.53	0.28	0.10	0.17	.....	.....	.....	.....	0.19	5.44	99.92	5.66	0.78	0.04	1.14	7.62	10
Hedjaz, Arabia	44.16	32.56	10.59	4.48	3.49	0.47	0.59	0.09	—	.....	.....	0.06	—	.....	3.03	99.52	12.98	0.94	—	1.61	15.53	11
Hendersonville, N.C.	48.09	29.88	11.76	2.30	2.22	1.00	0.10	0.61	.....	.....	.....	.....	.....	0.01	3.20	99.34	2.37	0.21	0.01	1.61	4.20	2
Hessle, Sweden	45.40	30.82	12.84	1.91	2.56	1.93	.....	.....	.....	.....	.....	.....	.....	Tr.	3.68	99.14	16.36	2.16	Tr.	0.18	18.70	2
Holbrook, Ariz.	41.89	29.08	16.96	4.29	2.40	Tr.	.....	.....	0.08	0.25	.....	.....	.....	.....	4.86	99.81	9.20	0.42	0.03	2.75	12.40	2
Homestead, Iowa	47.44	30.06	11.67	2.54	2.15	1.11	0.16	0.39	.....	0.10	.....	.....	0.57	0.23	3.46	99.88	13.68	1.19	0.09	2.28	17.24	12
Hvittis, Finland	59.30	33.17	0.38	2.21	2.01	1.80	0.46	0.81	.....	.....	.....	.....	.....	0.11	0.11	100.35	24.66	1.96	0.07	3.30	29.99	2
Indarch, Russia	47.84	22.87	14.95	2.64	1.55	0.17	.....	.....	0.74	.....	0.07	.....	3.75	0.63	4.44	99.65	19.30	0.95	0.02	5.10	25.37	2
Jelica, Yugoslavia	43.21	28.03	18.31	2.08	1.36	[0.13]		0.55	.....	0.21	.....	.....	.....	.....	5.24	99.12	6.12	0.83	0.05	2.58	9.58	2
Jerome, Kan.	35.26	22.99	23.16	1.89	1.40	0.69	0.30	0.62	1.89	Tr.	Tr.	Tr.	3.23	0.29	6.73	98.45	3.81	0.43	0.01	1.88	6.13	2
Johnstown, Colo.	50.76	27.12	11.40	2.37	2.65	0.33	0.10	1.03	.....	0.40	.....	.....	.....	0.02	3.27	99.45	0.55	.....	.....	.....	0.55	13
Juvinas, France	48.04	7.21	14.66	13.51	10.61	0.55	—	Tr.	0.11	.....	.....	0.57	.....	—	3.99	99.40	Tr.	.....	Tr.	0.06	0.06	2
Karoonda, Australia	35.54	25.99	21.94	5.81	2.70	0.74	0.27	0.51	.....	0.22	.....	—	0.14	0.20	6.55	100.81	2.95	0.02	.....	1.45	4.42	14
Kernouve, France	44.48	31.97	12.27	4.31	2.55	1.90	Tr.	Tr.	.....	.....	.....	.....	.....	.....	3.52	101.00	22.25	1.55	.....	2.15	25.95	2
Kesen, Japan	45.44	28.58	16.21	Tr.	2.78	0.56	0.76	.....	.....	0.34	.....	.....	.....	0.39	4.64	99.70	16.44	[2.32]		2.09	20.85	2

* Nomenclature according to A. L. Coulson, *Mem. Geol. Surv. India*, Vol. LXXV (1940), p. 1.

† Fe from FeO and Fe_2O_3.

‡ P from P and P_2O_5.

§ O from P_2O_5, FeO, and Fe_2O_3.

TABLE 1—*Continued*

Meteorite*	Silicate Phase															Iron-Nickel Phase					Ref.
	SiO_2	MgO	Fe†	Al_2O_3	CaO	Na_2O	K_2O	Cr_2O_3	NiO	MnO	CoO	TiO_2	H_2O	P‡	O§	Total	Fe	Ni	Co	S	Total	
Khairpur, India	57.35	31.06	2.84	2.37	2.33	1.08	0.13	0.55	—	.....	.....	—	0.27	Tr.	1.20	99.18	21.36	1.29	0.17	2.84	25.66	6
Klein-Wenden, Germany	46.11	33.00	7.53	5.23	3.95	0.39	0.53	0.87	.....	0.10	.....	.....	.....	0.03	2.16	99.90	23.90	2.37	.....	2.09	28.36	2
Knyahinya, Czechoslovakia	43.37	32.76	10.84	3.45	4.68	0.82	0.03	0.11	.....	0.85	.....	0.08	0.06	0.06	3.15	100.26	.....	.....	.....	.....	.....	15
Krahenberg, Germany	47.86	21.67	15.76	3.75	2.40	0.20	1.42	1.04	.....	0.91	.....	.....	.....	0.54	4.52	100.07	10.37	1.36	.....	2.35	14.08	2
Lanzenkirchen, Austria	46.64	27.59	11.48	4.98	1.94	2.68	0.32	0.51	.....	0.08	.....	0.08	0.06	0.10	3.32	99.78	11.48	1.10	0.06	1.90	14.54	16
Launton, England	46.41	29.10	12.44	2.81	2.44	1.18	0.11	0.36	0.14	0.34	.....	0.21	0.49	0.14	3.78	99.95	11.00	1.20	0.08	2.18	14.46	6
Limerick Co., Ireland	48.53	30.69	10.17	1.97	2.45	1.25	0.18	0.78	0.25	0.20	.....	.....	0.89	0.27	3.00	100.63	20.38	1.51	0.12	2.03	24.04	7
Linum, Germany	54.93	32.82	1.31	3.11	4.45	1.77	0.33	0.40	.....	0.26	.....	.....	0.15	0.10	0.37	100.00	19.06	0.71	.....	1.85	21.62	2
Lissa, Czechoslovakia	43.12	29.41	14.57	2.25	3.02	1.66	0.46	0.07	.....	1.12	.....	0.02	0.04	0.17	4.35	100.26	.....	.....	.....	.....	.....	15
Lixna, Latvia	46.00	31.64	13.11	3.18	Tr.	0.91	Tr.	0.56	.....	1.03	.....	.....	.....	0.18	3.75	100.36	16.95	1.71	.....	2.12	20.78	2
Long Island, Kan	37.61	23.99	18.66	3.25	1.48	0.26	0.03	6.68	0.81	Tr.	0.06	.....	1.60	0.06	5.35	99.84	2.60	0.67	0.04	1.90	5.21	2
Lundsgard, Sweden	45.51	29.29	12.61	3.32	1.72	1.75	0.53	0.73	0.06	.....	.....	.....	0.62	0.12	3.61	99.87	14.46	1.91	0.02	2.38	18.77	2
Lyubimovka, Russia	49.45	17.40	12.44	9.66	6.39	.....	.....	0.04	.....	c.72	.....	.....	.....	0.01	3.87	100.28	.....	.....	.....	0.18	0.18	17
Macibini, S. Africa	49.98	8.58	13.04	11.92	10.50	0.63	0.06	0.44	.....	0.52	.....	0.75	c.13	0.07	3.75	100.37	1.58	Tr.	—	0.21	1.79	18
Manbhoom, India	43.09	29.32	17.76	1.93	2.07	0.47	0.21	0.59	.....	0.08	.....	.....	.....	0.21	5.09	100.82	4.24	0.91	.....	1.70	6.85	2
Mangwendi, S. Rhodesia	44.00	26.03	16.88	4.72	1.68	0.50	c.12	0.49	0.37	0.15	.....	0.21	0.12	0.17	4.88	100.32	4.91	0.92	0.11	1.83	7.77	19
McKinney, Tex	43.21	30.43	6.55	15.15	1.88	.....	.....	0.08	0.50	0.24	.....	.....	.....	0.06	1.88	99.99	9.05	0.92	0.05	2.28	12.30	2
Merva, India	49.68	31.80	7.80	3.41	2.45	0.79	0.09	0.50	.....	0.24	.....	0.18	0.47	0.29	2.57	100.07	21.78	1.82	0.11	2.07	25.78	20
Misshof, Russia	45.06	24.92	11.87	0.37	.....	5.08	1.46	1.38	.....	5.63	.....	.....	.....	.....	3.40	99.17	18.80	1.35	.....	2.29	22.44	2
Mocs, Rumania	48.55	18.12	18.86	Tr.	3.16	1.36	0.24	1.20	.....	1.92	.....	.....	.....	0.47	5.40	99.28	7.93	1.38	Tr.	2.61	11.92	2
Modoc, Kan	48.32	28.96	13.08	2.70	1.91	0.48	Tr.	.....	.....	0.11	.....	.....	.....	0.05	3.75	99.36	6.56	0.68	0.03	1.38	8.65	2
Monroe, N.C	44.42	29.69	13.92	4.34	2.75	—	—	Tr.	.....	0.28	.....	.....	.....	.....	3.99	99.95	14.97	0.87	0.09	1.41	7.34	2
Moorleah, Tasmania	46.89	30.06	12.94	3.26	1.73	1.53	0.05	.....	.....	Tr.	.....	.....	0.06	Tr.	3.73	100.28	10.66	0.89	0.03	2.29	13.87	21
Mount Browne, Australia	43.72	29.33	12.21	2.85	2.81	1.47	0.30	0.03	.....	Tr.	.....	Tr.	.....	0.14	3.50	96.36	16.33	1.78	0.26	2.02	20.39	2
Nakhla, Egypt	49.01	12.02	16.18	1.74	15.19	0.41	0.14	0.33	.....	0.09	.....	0.38	0.07	.....	4.63	100.19	—	—	.....	0.06	0.06	2
New Concord, Ohio	46.74	24.24	21.52	0.31	0.02	[1.03]		.....	.....	Tr.	.....	.....	.....	.....	6.17	100.03	9.23	1.31	0.04	0.11	10.69	2
Ngawi, Java	47.13	16.87	20.72	0.86	2.90	3.01	0.50	0.39	1.73	Tr.	.....	.....	.....	.....	5.93	100.04	6.50	0.65	Tr.	2.08	9.23	2
Novo-Urei, Russia	41.84	37.91	10.99	0.64	1.48	.....	.....	1.01	.....	0.46	.....	.....	.....	0.02	3.15	97.50	5.25	0.20	.....	0.15	5.60	2
Oesede, Germany	47.95	31.33	8.37	3.65	3.99	1.78	0.59	.....	.....	.....	.....	.....	.....	.....	2.40	100.01	21.83	1.90	.....	2.11	25.84	22
Ojuelos Altos, Spain	47.70	30.25	10.51	3.30	2.97	0.91	0.27	.....	.....	.....	.....	.....	0.11	0.31	3.11	99.44	12.46	1.12	0.04	2.47	16.09	23
Olivenza, Spain	43.91	27.12	17.38	2.48	2.25	0.99	0.36	0.15	.....	0.16	.....	—	.....	0.15	5.03	99.98	4.59	0.41	0.02	2.19	7.21	24
Olmedilla de Alarcon, Spain	48.14	30.76	9.51	3.29	2.82	1.13	0.25	0.50	.....	0.34	.....	—	0.12	0.30	2.83	99.99	20.02	1.58	0.02	1.70	23.32	25
Perpeti, India	45.73	29.38	13.62	1.58	2.29	1.37	0.16	0.76	.....	0.32	.....	0.19	.....	0.24	4.48	100.12	10.89	1.18	0.05	2.28	14.40	26
Phu-Long, Cochin China	46.26	29.62	11.78	4.18	2.82	0.91	0.27	0.14	.....	0.17	.....	.....	0.08	0.20	3.44	99.87	18.68	1.21	0.02	1.65	21.56	27
Plantersville, Tex	48.09	29.63	11.82	2.74	2.48	1.32	0.15	0.04	0.01	0.23	—	—	.....	0.05	3.40	96.96	16.99	1.92	0.06	1.45	20.42	28
Prambachkirchen, Austria	46.37	28.25	11.66	4.67	2.70	1.40	0.28	0.06	.....	1.46	.....	0.14	.....	0.29	3.43	100.71	13.20	0.88	0.07	2.80	16.95	29
Rangala, India	46.82	28.86	13.67	2.66	2.42	0.71	0.16	0.34	.....	0.14	.....	0.09	0.04	0.23	4.04	100.18	11.52	1.00	0.14	1.86	14.52	30
Renca, Argentina	43.34	22.64	18.24	3.42	1.63	2.68	0.65	0.70	1.28	0.88	—	—	—	0.02	5.24	100.72	7.79	0.79	0.05	2.80	11.43	31
Rochester, Ind	46.55	30.33	14.74	0.11	2.77	0.65	.....	0.11	.....	.....	.....	.....	.....	.....	4.22	99.48	11.40	0.41	.....	1.09	12.90	2
Roda, Spain	51.29	27.59	11.80	2.91	1.45	0.39	0.32	0.65	.....	0.22	.....	.....	0.51	0.03	3.39	100.55	1.10	.....	.....	0.63	1.73	32
Rosebud, Tex	41.17	28.55	17.54	4.24	2.16	0.49	0.07	0.17	0.17	0.18	Tr.	—	0.08	0.08	5.06	99.96	14.32	1.15	0.05	1.25	16.72	33
St. Christophe, France	47.08	31.00	12.71	2.57	1.81	0.61	0.22	0.47	.....	.....	.....	.....	.....	.....	3.64	100.11	12.17	1.67	0.11	2.52	16.47	2
St. Marks, S. Africa	57.78	27.51	7.63	0.97	2.20	1.28	0.35	.....	.....	1.06	.....	.....	.....	0.08	2.18	101.04	26.44	1.84	0.21	5.26	33.75	2
St. Michel, Finland	46.63	29.03	12.33	3.91	1.94	1.56	0.15	0.66	.....	0.48	.....	0.02	.....	0.09	3.53	100.33	11.71	1.16	0.13	2.22	15.22	2
Saline, Kan	41.46	26.09	19.15	2.05	2.27	0.29	0.09	1.40	0.83	.....	0.08	.....	1.38	0.06	5.49	100.64	7.89	0.95	0.04	1.65	10.53	2
Salt Lake, Utah	46.61	29.76	12.28	2.53	2.42	1.10	0.08	0.54	.....	.....	.....	.....	1.22	0.26	3.60	100.40	19.18	1.38	0.10	2.00	22.66	2
Santa Isabel, Argentina	47.22	23.32	15.79	3.37	2.92	0.76	0.27	0.76	1.15	0.45	Tr.	Tr.	.....	0.16	4.59	100.76	8.37	0.30	0.02	2.83	11.61	34

TABLE 1—*Continued*

Meteorite*	Silicate Phase															Iron-Nickel Phase					Ref.
	SiO_2	MgO	Fe†	Al_2O_3	CaO	Na_2O	K_2O	Cr_2O_3	NiO	MnO	CoO	TiO_2	H_2O	P‡	O§	Total	Fe	Ni	Co	S	Total	
Sazovice, Czechoslovakia	45.87	27.73	13.84	3.68	2.83	1.03	.....	.....	.....	0.83	.....	0.07	0.10	0.05	3.97	100.00	.....	.....	.....	.....	.....	35
Schoneberg, Germany	48.44	16.67	16.24	6.72	2.79	2.66	0.88	0.49	.....	.....	.....	.....	.....	0.43	4.65	99.97	13.77	1.47	.....	1.93	17.17	2
Sharps, Va.	43.24	28.69	14.46	3.33	3.05	0.94	0.21	.....	0.48	0.36	—	.....	.....	0.17	4.17	99.15	.....	.....	.....	0.58	0.58	36
Shelburne, Canada	45.11	30.20	13.56	2.47	2.01	0.84	0.25	0.71	.....	0.14	.....	.....	.....	0.07	3.89	99.25	10.70	0.78	0.04	1.61	13.13	2
Soko-Banja, Yugoslavia	45.72	28.27	15.20	2.34	2.12	0.94	0.08	0.40	.....	0.06	.....	0.11	0.19	0.20	4.42	100.05	6.34	1.06	0.09	1.81	9.30	6
Stalldalen, Sweden	47.71	30.94	10.60	2.82	2.15	0.83	0.20	0.53	0.27	0.33	.....	.....	.....	0.19	3.06	99.72	21.10	1.61	0.17	2.27	25.15	2
Stannern, Czechoslovakia	48.37	7.20	14.88	11.29	10.45	0.75	0.13	0.35	0.25	.....	Tr.	0.41	0.30	0.11	4.29	98.79	0.55	.....	.....	0.31	0.86	2
Strathmore, Scotland	47.38	29.33	12.15	3.02	2.16	1.06	0.13	0.52	.....	0.34	.....	0.16	0.20	0.20	3.59	100.24	11.31	1.26	0.05	2.29	14.91	37
Tabor, Czechoslovakia	43.89	32.10	11.55	2.91	3.82	1.25	0.09	0.10	.....	0.71	.....	0.09	0.10	0.14	3.43	100.18	.....	.....	.....	.....	.....	15
Tataovine, Southern Tunis.	55.16	27.53	11.94	0.62	0.76	.....	—	0.35	—	0.26	.....	0.19	0.14	.....	3.42	100.37	0.22	.....	.....	0.13	0.35	38
Tennasilm, Esthonia	45.29	25.91	15.86	2.97	1.59	[2.61]		1.07	.....	.....	.....	.....	.....	0.08	4.54	99.92	11.77	1.67	.....	0.63	14.07	2
Tilden, Ill.	44.72	24.20	18.79	4.00	0.99	0.58	—	.....	1.07	.....	0.04	.....	0.03	0.12	5.42	99.96	6.38	0.31	0.02	1.15	7.86	39
Tjerebon, Java	43.22	26.06	20.12	.....	0.97	0.91	0.52	0.74	0.16	0.57	.....	.....	.....	.....	6.05	99.32	14.21	1.49	.....	1.99	17.69	40
Travis County, Tex.	46.09	28.77	12.84	2.80	2.30	1.16	0.13	0.54	0.54	Tr.	.....	.....	0.87	0.32	3.78	100.14	1.80	0.22	0.01	0.91	2.94	2
Tuantuc, Cochin China	46.36	28.94	13.33	3.41	2.29	0.77	0.24	0.16	.....	0.24	.....	.....	0.07	0.25	3.90	99.96	9.93	0.78	0.02	2.24	12.97	27
Utrecht, Holland	45.93	28.42	14.05	2.61	1.73	1.62	0.17	0.51	.....	.....	.....	.....	.....	Tr.	4.02	99.06	11.07	1.24	.....	1.94	14.25	2
Wittekrantz, S. Africa	46.67	28.83	13.18	2.88	2.41	1.32	0.16	0.41	.....	0.17	.....	0.19	0.18	0.14	3.82	100.36	9.85	0.75	0.02	1.26	11.88	2
Zebrak, Czechoslovakia	43.04	32.14	12.45	2.85	3.05	1.54	0.33	0.08	.....	1.07	.....	0.05	0.06	0.09	3.65	100.40	.....	.....	.....	.....	.....	15
Zemaitkiemis, Lithuania	48.80	27.04	11.33	3.73	2.78	2.49	0.31	.....	.....	0.27	.....	0.06	.....	0.27	3.36	100.44	14.74	0.80	.....	1.90	17.44	41

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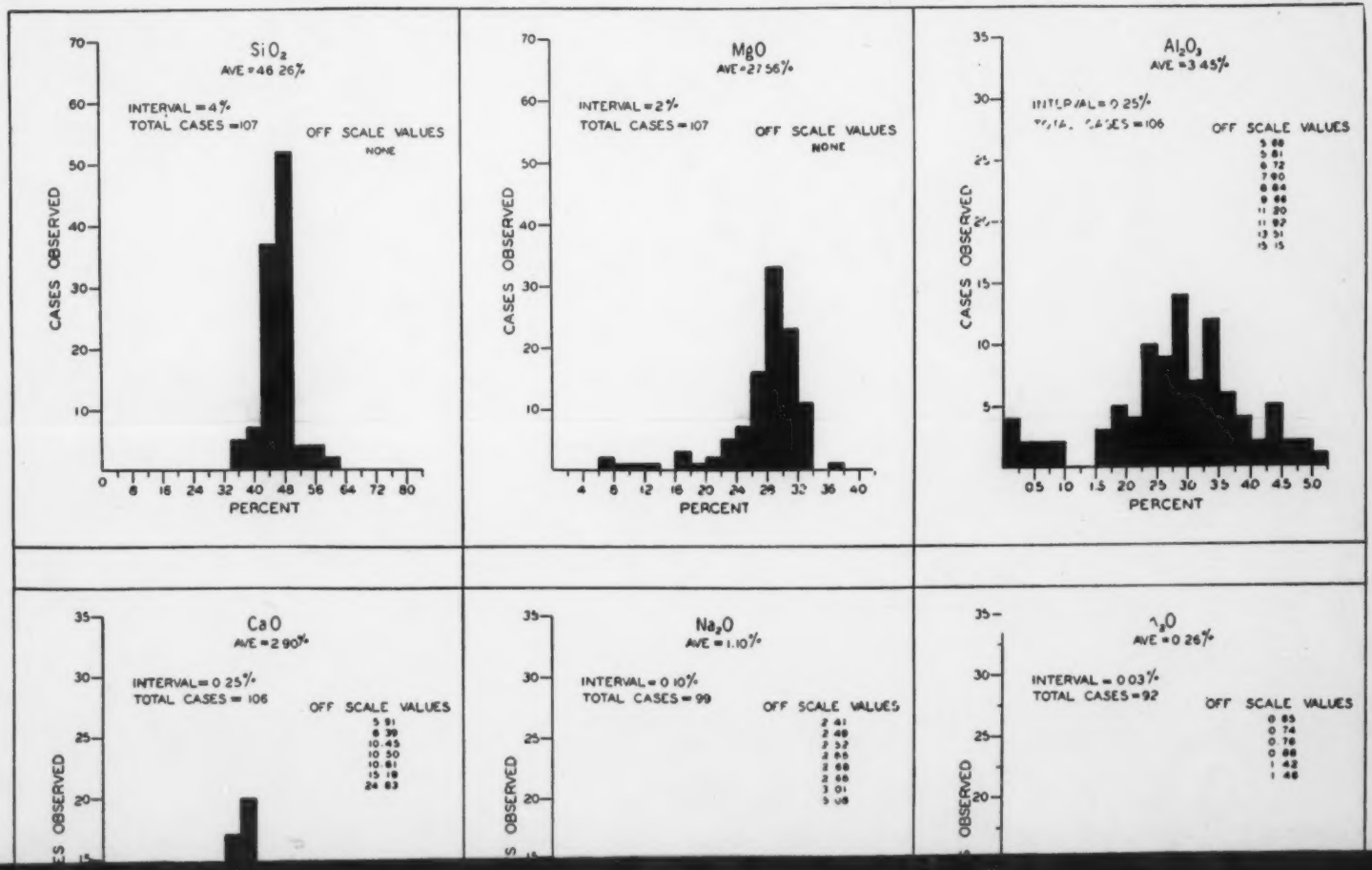
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INTERVAL = 4%
TOTAL CASES = 107
OFF SCALE VALUES
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CASES OBSERVED
PERCENT
MgO
AVE = 27 56%
INTERVAL = 2%
TOTAL CASES = 107
OFF SCALE VALUES
NONE
CASES OBSERVED
PERCENT
Al_2O_3
AVE = 3 45%
INTERVAL = 0 25%
TOTAL CASES = 106
OFF SCALE VALUES
5 68
5 81
6 72
7 90
8 84
9 66
11 20
11 92
13 51
15 15
CASES OBSERVED
PERCENT
CaO
AVE = 2 90%
INTERVAL = 0 25%
TOTAL CASES = 106
OFF SCALE VALUES
5 91
6 39
10 45
10 50
10 61
15 18
24 83
Na_2O
AVE = 1 10%
INTERVAL = 0 10%
TOTAL CASES = 99
OFF SCALE VALUES
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2 48
2 52
2 65
2 68
2 68
3 01
3 08
AVE = 0 26%
INTERVAL = 0 03%
TOTAL CASES = 92
OFF SCALE VALUES
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0 74
0 76
0 88
1 42
1 46

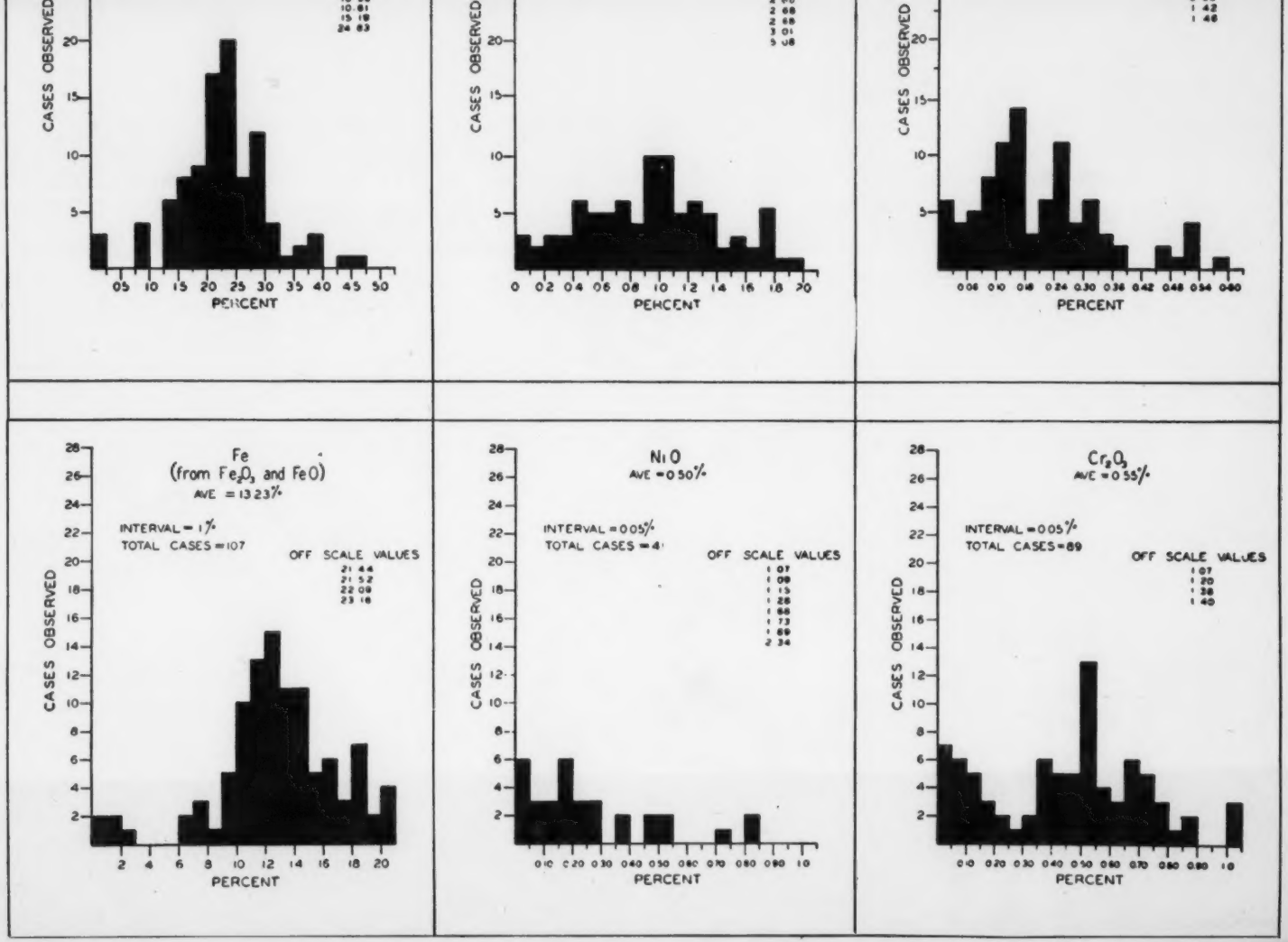

FIG. 1.—Frequency-distribution curves for the major constituents of the silicate phase of stony meteorites

FIG. 1.—Frequency distribution curves for the major constituents of the silicate phase of stony meteorites

Merrill's compilation, and the balance have been selected from the literature accumulated since 1916. The data have been broken down in the following manner: The metallic iron, nickel, and cobalt and the sulphur present in each meteorite have been subtracted from the gross analysis. The balance, representing the silicate phase, has been adjusted so that the sum of the constituents equals 100 per cent. Wherever possible the direct analysis of the silicate phase, freed magnetically from the metallic and sulphide phases, was used, in which case no numerical adjustments were necessary.

Figure 1 shows frequency-distribution curves for the major constituents. The number of observed cases lying within a designated percentage interval is plotted against the location of the interval. The interval chosen in each case is approximately 10 per cent of the arithmetic mean. It can be seen that the two most abundant constituents, silica and magnesia, are fairly well defined and that for the less abundant constituents the spread is considerably greater.

Table 2 shows the arithmetic mean of each constituent, together with the standard deviation σ, where

$$\sigma = \sqrt{\frac{\Sigma \Delta_i^2}{n-1}};$$

Δ_i = Deviation of the abundance of a given constituent in a given meteorite from the arithmetic mean;

n = Number of meteorites studied.

The standard deviation is essentially a measure of the "spread" of an element or the range of concentrations in which it is most prevalent. If a sufficient number of cases is available for the data to be representative, σ should be fairly independent of n.

TABLE 2

AVERAGE COMPOSITION BY WEIGHT OF THE SILICATE PHASE OF STONY METEORITES

Constituent	Per Cent	Standard Deviation	Precision	Observed Cases
SiO_2	46.26	4.18	±0.40	107
MgO	27.56	5.44	±0.53	107
Fe*	13.25	4.39	±0.42	107
Al_2O_3	3.45	2.92	±0.28	106
CaO	2.90	2.96	±0.29	106
Na_2O	1.10_3	0.75	$\pm 0.07_5$	99
K_2O	0.25_3	0.25	±0.026	94
Cr_2O_3	0.54_2	0.73	$\pm 0.07_8$	89
	0.47_2	0.33	$\pm 0.03_5$	88
MnO	0.41_3	0.71	$\pm 0.07_8$	82
	0.35_1	0.36	$\pm 0.03_9$	81
NiO	0.50_2	0.58	$\pm 0.09_1$	41
TiO_2	0.17_3	0.37	$\pm 0.05_3$	48
	0.12_5	0.16	$\pm 0.02_3$	47
CoO	0.027	0.046	±0.011	16
H_2O	0.58_3	1.00	$\pm 0.14_3$	49
P	0.17	0.13	±0.14	85
O†	3.84	1.29	±0.12	107

* From Fe_2O_3 and FeO.
† From Fe_2O_3, FeO, and P_2O_5.

The precisions of the arithmetic means are given in the fourth column of Table 2 and have been calculated, using the relationship

$$M = \pm \frac{\sigma}{\sqrt{n}},$$

where M is the measure of precision. In the cases of the three constituents—titanium oxide, chromium oxide, and manganese oxide—the arithmetic means are affected markedly by the decision as to whether to include in each case one abnormally high value. For these cases the means, together with the deviations and precisions, were calculated both with and without the abnormal value.[4]

In Table 3 the composition has been broken down in terms of elements and expressed in three ways: weight per cent, atomic per cent, and (after Goldschmidt) in terms of atoms per 100 atoms of silicon. In the cases of titanium, chromium, and manganese the averages given are the means of the high and low values.

[4] The abnormal values are as follows: Angra dos Reis, Brazil, 2.42 per cent TiO_2; Long Island, Kansas, 6.68 per cent Cr_2O_3; Misshof, Russia, 5.63 per cent MnO.

It can be seen that in the most favorable case, that of silicon, the mean is probably correct to within 1 per cent of the mean. In the least favorable case, that of titanium, the mean is probably correct to within 35 per cent of the mean.

If one plots the standard deviation divided by the arithmetic mean against the logarithm of the arithmetic mean for each of the metallic oxides, an interesting trend becomes evident. Figure 2 indicates that, as the concentration decreases, the standard deviation increases rather markedly. While at least some of the observed spread can be attributed to experimental error, it seems clear that the major portion of the spreads, shown in the plot, actually occur in nature. While the curve, arbitrarily drawn through the points, should not be regarded as a precise relationship, Figure 2 indicates that in the case of the minor constituents (constituents present to an extent less than 100 parts per million) a very large number of falls must be analyzed if arithmetic means possessing fair degrees of precision are to be

TABLE 3

AVERAGE COMPOSITION OF THE SILICATE PHASE OF STONY METEORITES BY ELEMENTS

Element	Weight Per Cent	Atomic Per Cent	Atoms per 100 Atoms of Si
Oxygen	43.10 ±1.00	58.09 ±1.36	350 ±11
Silicon	21.61 ±0.19	16.60 ±0.15	100
Magnesium	16.62 ±0.32	14.73 ±0.28	88.7 ± 2.5
Iron	13.25 ±0.42	5.11 ±0.16	30.8 ± 1.3
Calcium	2.07 ±0.21	1.11 ±0.11	6.70 ± 0.74
Aluminum	1.83 ±0.15	1.46 ±0.12	8.82 ± 0.81
Sodium	0.81_8 ±0.056	0.76_8 ±0.05_2	4.62 ± 0.36
Nickel	0.39_4 ±0.07_2	0.14_5 ±0.02_5	0.87 ± 0.16
Chromium	0.36_2 ±0.06_2	0.15_6 ±0.02_5	0.90 ± 0.16
Manganese	0.31_1 ±0.07_0	0.12_2 ±0.02_5	0.74 ± 0.16
Potassium	0.20_9 ±0.02_1	0.11_5 ±0.01_1	0.69_3 ± 0.07_5
Titanium	0.098±0.032	0.04_3 ±0.01_4	0.26_0 ± 0.09_0
Phosphorus	0.16_6 ±0.01_4	0.11_6 ±0.01_0	0.69_6 ± 0.06_5
Hydrogen	0.066±0.016	1.40 ±0.35	8.4 ± 2.2
Cobalt	0.022±0.009	0.0079±0.0033	0.048± 0.020

obtained. For example, extrapolation of the curve in Figure 2 back to 10 parts per million (0.001 per cent) indicates that, for a constituent present at that average concentration, some 500 falls should be analyzed (either collectively or separately) if the arithmetic mean is to be precise to 10 per cent.

no greater a precision than 25–35 per cent on a purely statistical basis. Such a precision, however, would be correct only if no error is inherent in the analytical work. When one takes into consideration the difficulties of precise analysis, it seems only reasonable to suppose that most of the work done to date on the

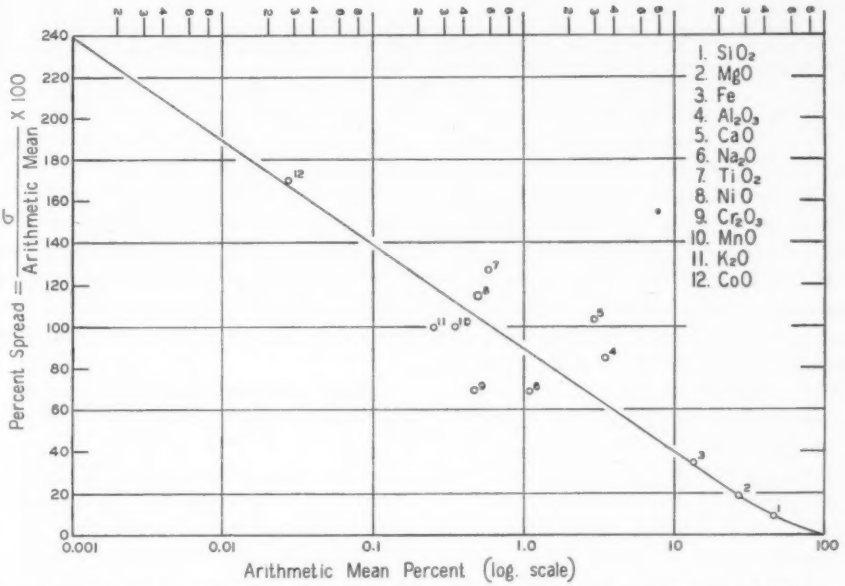

FIG. 2.—The variation of the spread with the arithmetic mean for the major constituents of the silicate phase of stony meteorites.

Thus it would appear that in the extensive work of the Noddacks[5] on the minor constituents of stony meteorites (a composite mixture of 42 falls was used), the results are probably valid to

minor constituents of stony meteorites has a precision of probably no greater than a factor of 2.

ACKNOWLEDGMENTS.—The authors are indebted to Professors T. Barth and Jerome Fisher, of the Department of Geology of the University of Chicago, and to Dr. S. K. Roy, of the Chicago Natural History Museum, for many stimulating conversations on the general subject of this paper.

[5] I. and W. Noddack, "Die Häufigkeit der chemischen Elemente," *Naturw.*, Vol. XVIII (1930), p. 757; "Die geochemischen Verteilungskoeffizienten der Elemente," *Svensk kem. Tidssk.*, Vol. XLVI (1934), p. 173.

THE LONGVIEW MEMBER OF THE KINGSPORT FORMATION

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ABSTRACT

A detailed stratigraphic study of the Longview member of the Kingsport formation of the Knox dolomite group has been undertaken because of the economic importance of the Knox dolomite to zinc mining in the southeastern United States.

An excellent opportunity presented itself to the author to study and correlate detailed sections in underground exposures of zones about which little could be learned from surface exposures because of the scarcity of outcrops, the variation in lithology, and the lack of fossils.

INTRODUCTION

The Mascot properties of the American Zinc, Lead, and Smelting Company are in Knox County, Tennessee, 13 miles northeast of Knoxville, about equidistant between U.S. Highways 11*E* and 11*W*. The data for this paper were gathered from a study of underground exposures in the Mascot mines and from surface outcrops on the company property and near-by localities.

The mineralized district lies within the Tennessee section of the Valley and Ridge Province, a southwest extension of the Shenandoah Valley of Virginia. This part of the Valley and Ridge Province is approximately 50 miles wide, being bounded on the northwest by the Cumberland Plateau and on the southeast by the Great Smoky Mountains. Topographically, it is a region of ridges with narrow intervening valleys. The ridges are formed of resistant chert and sandstone, and the valleys of less resistant limestone, dolomite, and shale, The ridges are largely wooded. The valleys are used for agriculture and pasture land.

HISTORY OF ZINC MINING IN TENNESSEE

Zinc mining in eastern Tennessee dates back to the middle of the last century. Oxide and sulphide ores occur at many places. Where conditions were favorable for their formation, carbonate ores occurred in the outcrop, from which the earliest production was derived.

Tennessee first came to general attention as an important producer of zinc ores in 1913, when the Embree Iron Company shipped its first oxidized ores from Embreeville and the American Zinc Company its first sulphide ores from Mascot.

The Mascot–Jefferson City zinc district extends from Mascot to Jefferson City, about 30 miles northeast of Knoxville. The principal producers of sulphide ores at the present time are the American Zinc Company mines at Jefferson City, New Market, and Mascot; and the Universal Exploration Company mine at Jefferson City. The approximate daily production is 5,000 tons of ore, grading 2.5–5 per cent zinc.

Zinc ores occur at various horizons in the Knox dolomite. The position of the most productive ores at Mascot has been a zone 97 feet thick, occurring some 150 feet above the top of the Longview member; whereas the ore mined at New Prospect, 25 miles northwest of Mascot, is 2,500 feet stratigraphically lower. Since the Longview occupies an intermediate position and in some localities is known to contain zinc minerals, a detailed description of it should be useful.

GENERAL GEOLOGY OF THE DISTRICT

The formations of the district are of limestones, dolomites, and shales, ranging in age from Cambrian to Carboniferous.[1] The strata have been intensely folded and faulted, the structures being essentially parallel and trending northeasterly with the valley. The prevailing rock dips in this region of Appalachian overturning are to the southeast. Displacements along some of the overthrusts amount to as much as 6–8 miles. The folding and faulting have allowed the unequally resistant strata to form alternate ridges and valleys and to outcrop in a succession of belts.

The Knox dolomite, which ranges in thickness from 2,800 to 3,200 feet, is the most important and the most widespread of the valley rocks. It is made up of at least four formations, which are so similar in appearance that very close study is required to distinguish them. These are the Conococheague, Chepultepec, Kingsport, and Mascot. Their stratigraphic position, subdivisions, and approximate thicknesses are shown in the accompanying generalized section (Table 1).

The Kingsport formation of the Knox group is composed of 550–625 feet of limestone and dolomite. The dolomite varies in color from very light to dark gray and in texture from fine grained to very coarse grained. The limestone is

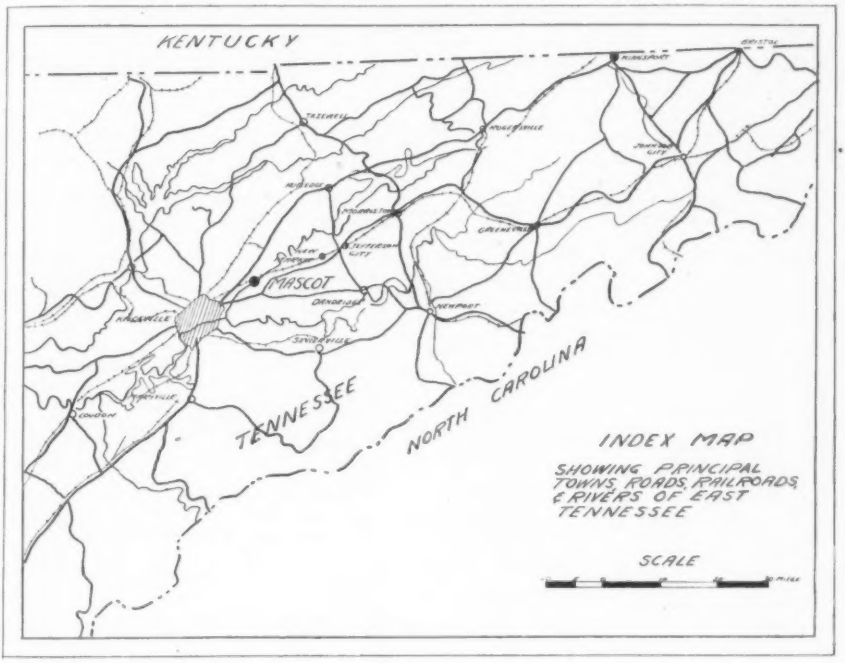

FIG. 1

[1] M. H. Newman, "The Mascot–Jefferson City Zinc District, *XVI Int. Geol. Cong. Guidebook 2* (1932), pp. 152–65.

dove to brown in color and is fine grained. Numerous arenaceous layers and shale seams are present, but few are of very great thickness. Many bands of chert nodules and chert layers are scattered throughout the section. The lowest divison of the Kingsport is the Longview dolomite member.

STRATIGRAPHY OF THE LONGVIEW DOLOMITE

In older publications the portion of the Knox dolomite represented by the Longview dolomite member was called "Nittany" because it was believed to correlate with a formation bearing that name in Pennsylvania. Recent workers, how-

TABLE 1

GENERALIZED SECTION, MASCOT–JEFFERSON CITY ZINC DISTRICT, EASTERN TENNESSEE

Age		Formation Name			Thickness in Feet	Character of Rocks
Ordovician	Chazyan	Lenoir and Mosheim limestone (Chickamauga)			250±	Dark gray (Lenoir) and dove bird's-eye (Mosheim) limestone
Ordovician	Canadian	Knox dolomite	Mascot (Cotter-Powell)		500–700	Light and dark-gray dolomite and limestone moderately cherty; limestone most abundant in Jefferson City area; base marked by chert-matrix sandstone; rarely mineralized
Ordovician	Canadian	Knox dolomite	Kingsport (Jefferson City)	Upper dolomite	184±	Very light- to medium light-gray dolomite, moderately cherty; extremely fine-grained with small amount semicrystalline and crystalline dolomite; base marked by 5-foot bed of very dark dolomite
Ordovician	Canadian	Knox dolomite	Kingsport (Jefferson City)	Middle limestone	210±	Brown limestone, which in many places has been altered to crystalline dolomite; some some light- to dark-gray dolomite
Ordovician	Canadian	Knox dolomite	Kingsport (Jefferson City)	Longview dolomite	150±	Dolomite, light gray to nearly white; fine-grained to semicrystalline dolomite
Cambrian	Ozarkian	Knox dolomite	Chepultepec		500±	Mostly dolomite at Mascot; light- and dark-gray dolomite and limestone in Jefferson City area; very cherty; heavy sandstone at base
Cambrian	Ozarkian	Knox dolomite	Conococheague		500±	Dark sugary and light-gray, fine-grained cherty dolomite; some limestone in Jefferson City area
Cambrian	Croixan	Maynardsville limestone			60–250+	Light-blue to dark-gray laminated limestone and dolomite
Cambrian	Croixan	Nolichucky shale			550–750	Dark-gray, greenish, and yellow shales and thin limestones

ever, have replaced the name Nittany with that of "Longview," which was first used by Charles Butts[2] in describing rocks in Alabama which are now believed to correlate with this zone in Tennessee.

The Knox dolomite has been described by various authors, but, with the exception of the descriptions of the Kingsport formation by C. R. L. Oder and W. H. Miller[3] and by J. Crawford,[4] practically all the descriptions have dealt with surface outcrops involving, for the most part, poor exposures of the Longview.

The detailed stratigraphys and the true relations of the Longview have heretofore been poorly understood because surface exposures of the rocks in the district are few. Furthermore, fossils are sparse and difficult to find, and the ones that can be found occur in residual chert. Lithology appears to be more practicable than fossils for establishing the the Longview as a mappable unit and for understanding its nature and relation to the Kingsport formation. Since the parts of this unit seen in underground workings at Mascot are more completely exposed than in any other locality in the

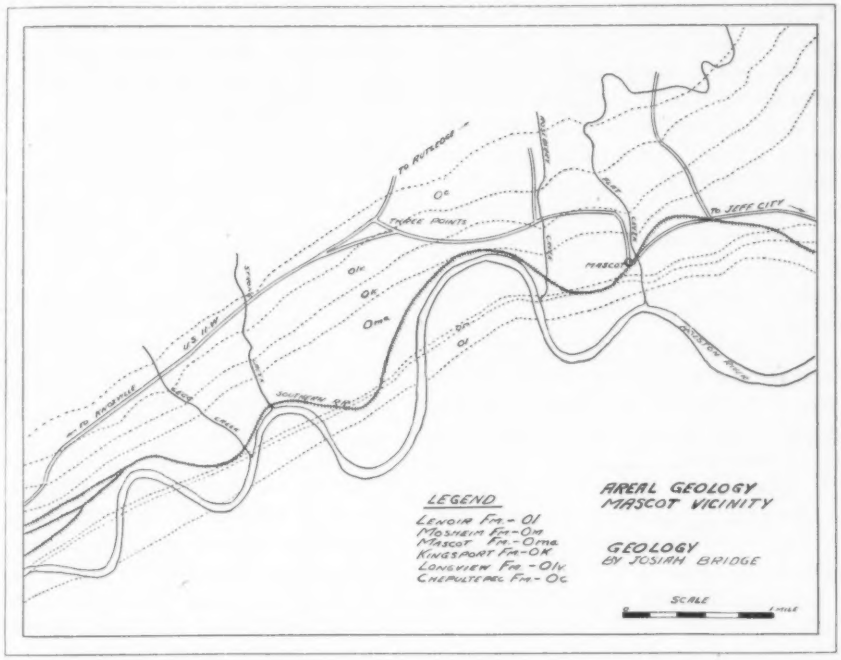

FIG. 2

[2] "Geology of Alabama, the Paleozoic Rocks," *Ala. Geol. Surv. Spec. Rept. 14* (1926).

[3] "Stratigraphy of the Mascot–Jefferson City Zinc District," *A.I.M.E. Tech. Pub. 1818* (1945), pp. 1–9.

[4] "Structural and Stratigraphic Control of Zinc Deposits in East Tennessee," *Econ. Geol.*, Vol. XL, No. 6 (1945), pp. 412–15.

valley, except in the section at Thorn Hill, the mine sections can best furnish the detailed descriptions.

The Longview member of the Mascot district is composed of 145 feet of light- to very-light-gray dolomite, for the most part in fine-grained layers, although some layers are so coarsely crystalline that they resemble marble in appearance. The rock approaches theoretical dolomite in composition and may have been altered from fine-grained limestone. Some layers show a replacement of the limestone by crystalline dolomite within only a few feet in underground exposures. Arenaceous zones of medium-sized frosted quartz grains disseminated in dolomite are numerous, although most of the silica content occurs as interstitial filling. Chert nodules and chert layers are scarce.

The late Mark H. Newman devised the method of measuring and naming the beds in the mines at Mascot and described parts of the ore-bearing zones. His method utilized numbers to denote the thickness of beds above and below the footwall sand, which he designated the "0" bed. These numbers also were used as the name of the individual bed.

Oder and Miller's studies did not include any of the Longview. In fact, they did not cover any of the beds between the top of the Longview and the layer which they termed the "−64 bed" at Mascot. Newman recognized the presence of sands below −64 bed, but he failed to describe any part of that zone.

SUMMARY

Former workers have considered the thickness of the Longview in the Mascot locality to be 250 feet. The writer's examination of the strata included in that measurement shows that it includes about 100 feet of crystalline dolomite and layers of true limestone, which belong in the overlying middle limestone member of the Kingsport.

The writer has found no radical change in sedimentation or other evidence of unconformity at the top of the Longview in this area. The lithology of that unit is more like that of the overlying Kingsport than that of the underlying Chepultepec beds. Therefore, it is here considered to be a member of the Kingsport formation, which the author believes consists of three members as follows:

1. The upper dolomite member; 184 feet thick; principally light-gray, fine-grained dolomite, with a few thin layers of limestone
2. The middle limestone member; 210 feet thick; composed of brown limestone, which in many places has been altered to crystalline dolomite with some layers of light- to dark-gray, fine-grained dolomite
3. The Longview dolomite member; 145 feet thick; composed of light-gray to nearly white, fine-grained dolomite, with a few beds of light-gray dolomite of medium crystalline texture.

The crystalline dolomite in the lower part of the middle member of the Kingsport formation is referred to locally as "recrystalline dolomite." When one follows this zone along the strike of mine sections, the crystalline texture may be seen to grade into unaltered, fine-grained, dove-colored limestone. In some localities dolomitization and the development of a crystalline texture are practically complete, with only a few remnants of unaltered limestone remaining. The evidence noted in this zone is sufficiently conclusive to state that the crystalline texture is a result of alteration.

Whether or not any limestone occurs in the Longview has been a matter of controversy. It is certainly present in this zone in the Thorn Hill section, but only one small occurence of actual lime-

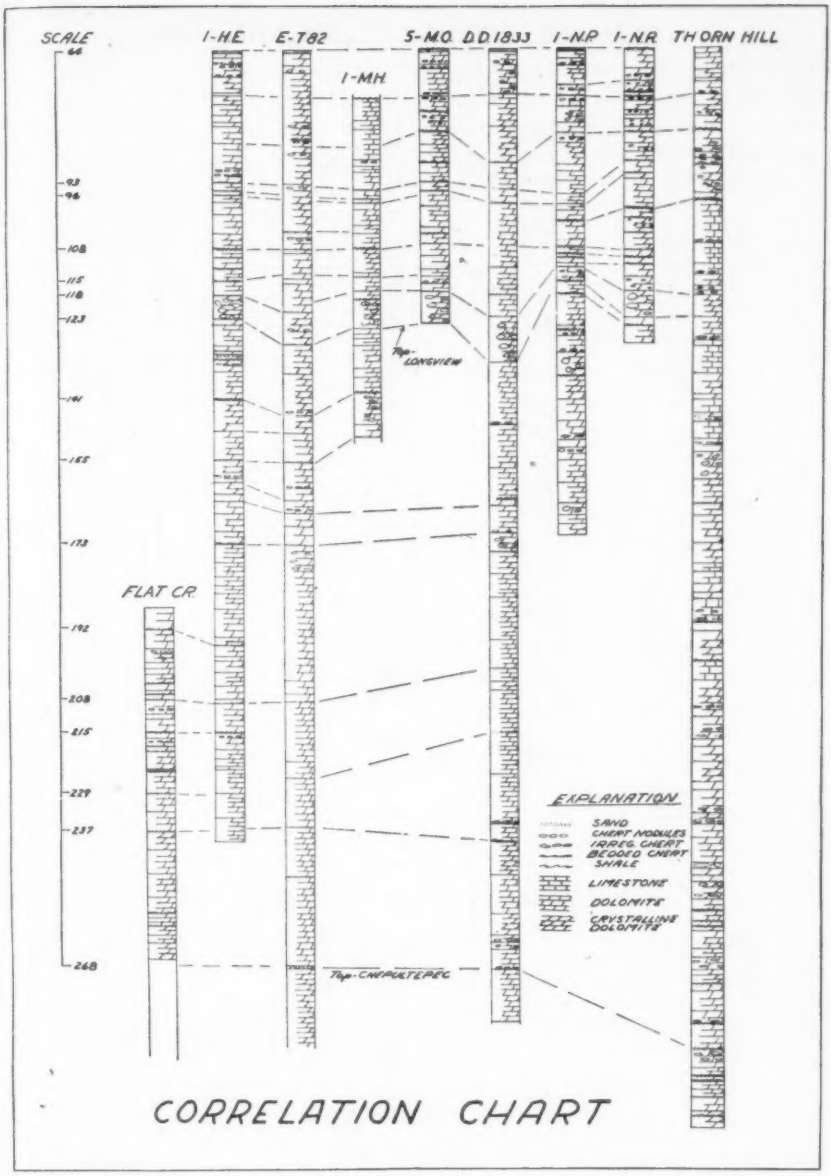
SCALE
64
93
96
108
115
118
123
141
155
173
192
208
215
229
237
268
I-HE
E-T82
I-MH
5-MO
DD1833
I-NP
I-NR
THORN HILL
Top LONGVIEW
FLAT CR.
Top CHEROKEE EC.
EXPLANATION
SAND
CHERT NODULES
IRREG. CHERT
BEDDED CHERT
SHALE
LIMESTONE
DOLOMITE
CRYSTALLINE DOLOMITE
CORRELATION CHART

FIG. 3

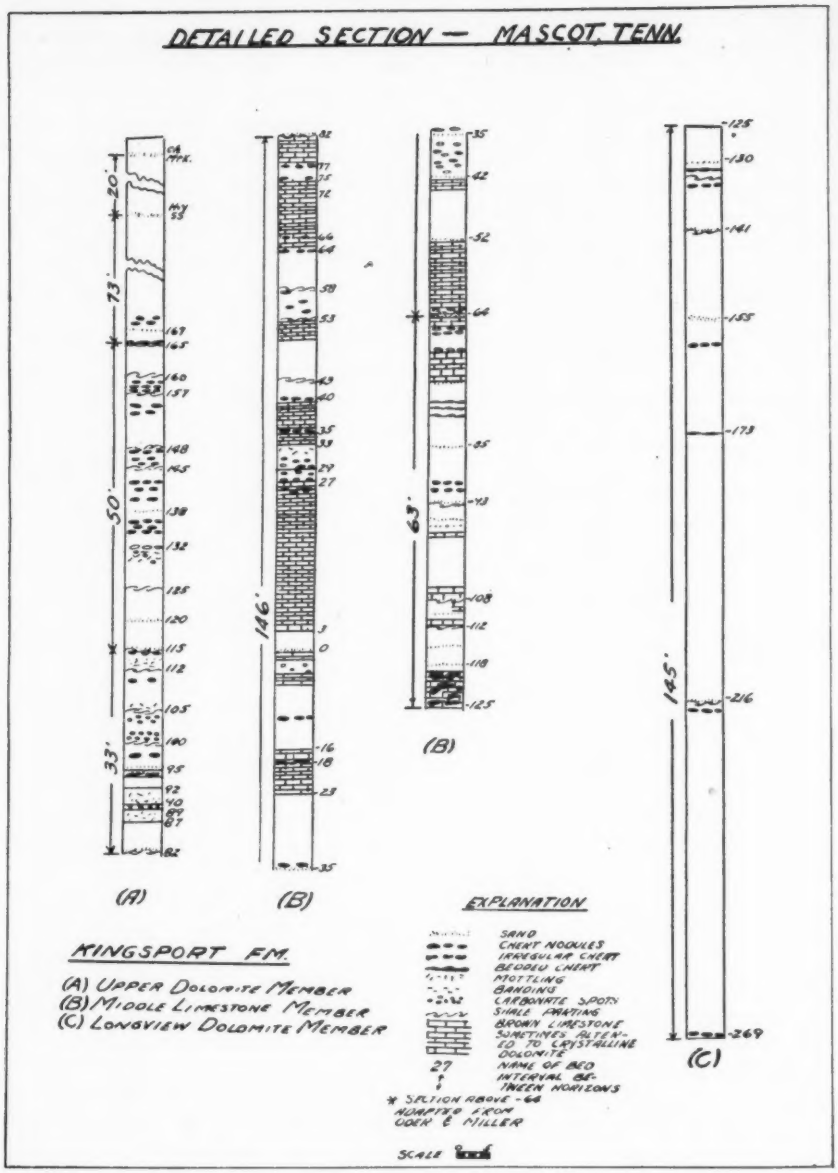
DETAILED SECTION — MASCOT, TENN.
(A)
(B)
(C)
KINGSPORT F.M.
(A) UPPER DOLOMITE MEMBER
(B) MIDDLE LIMESTONE MEMBER
(C) LONGVIEW DOLOMITE MEMBER
EXPLANATION
SAND
CHERT NODULES
IRREGULAR CHERT
BEDDED CHERT
MOTTLING
BANDING
CARBONATE SPOTS
SHALE PARTING
BROWN LIMESTONE
SOMETIMES ALTERED TO CRYSTALLINE DOLOMITE
NAME OF BED
INTERVAL BETWEEN HORIZONS
* SECTION ABOVE -66 MODIFIED FROM COER & MILLER
SCALE

FIG. 4

stone has been found in the Longview at Mascot, although some of the crystalline dolomite occurring in this member in the latter locality represents altered limestone.

Some workers have thought that chert is far more abundant in the Longview than in other parts of the Kingsport formation. This opinion seems to be due to the heavy chert residuum usually found along the surface outcrop of this zone. However, the writer has found that this is not always true. If the limestone and crystalline dolomite members are placed in the Longview, because of the presence of *Lacanospira*, the Longview may be considered to be as cherty as the middle member; but if the limestone beds are placed in the middle member of the Kingsport, the amount of chert in the Longview, other than interstitial silica, is small in quantity.

ACKNOWLEDGMENTS.—The writer wishes to express his appreciation to the American Zinc Company, especially to Mr. Harley A. Coy, general superintendent, for permission to do this work and to Dr. Charles R. L. Oder, chief geologist, for advice and many helpful suggestions.

ON THE GEOCHEMICAL CYCLE OF FLUORINE

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ABSTRACT

The average concentration of fluorine in the earth's crust is about 800 gm. per ton. In basalts and gabbros it is present as apatite; in granites, gneisses, and metamorphic rocks it is contained in mica, hornblende, and fluorspar. Much fluorine enters into the magmatic gas phase, which has an acid reaction. Great amounts of fluorine are therefore exhaled by volcanic eruptions. If the magmatic gas phase has opportunity to react with the wall rock, it turns alkalic, and fluorine is extracted (as phosphate?) before it reaches the surface; alkaline hot springs carry no magmatic fluorine. Of the fluorine dissolved by the weathering processes, only 0.2 per cent is found in the ocean. Fluorine is extracted from inland waters and from the sea, forming mineral deposits with a composition resembling fluorapatite.

THE AVERAGE FLUORINE CONCENTRATION IN THE LITHOSPHERE

Geochemistry is concerned with the chemical composition of the earth's crust as a whole, with the differences in composition between the individual rocks and minerals, and with chemical changes occurring in the rocks. It traces the geochemical cycle of each element and explains reactions taking place at the various depths, e.g., how some elements in certain stages of their geochemical cycle concentrate in ore deposits or salt layers, while others during their entire cycle occur as "disperse" elements in very small concentrations.

Fluorine is one of these "disperse" elements, never concentrating in large mineral deposits, and, in addition, it has been difficult to catch analytically in rocks and minerals.

In 1920, F. W. Clarke[1] on very scanty data estimated fluorine to constitute about 200 gm. per ton of the earth's crust. Four years later F. W. Clarke and H. S. Washington[2] were so discouraged by the uncertainty of the analytical methods available for determining fluorine that they virtually neglected the data of the analysts and estimated the amount of fluorine indirectly, on the assumption that the chief carrier of fluorine in igneous rocks was the mineral apatite. Consequently, they argued, the average fluorine should be about one-tenth that of the phosphoric acid, i.e., 300 gm. per ton. This figure is now currently used in geochemical writings.

But with the publication of H. H. Willard and O. B. Winter's[3] reliable method for determining fluorine, E. S. Shepherd[4] at the Geophysical Laboratory started a long series of analyses aiming to establish the significance of fluorine in geochemistry.[5] He found that about 400 gm. per ton is characteristic of plutonic rocks, although alkaline rocks seemed to average higher. His data are interesting also in showing that fluorine does not, as had been suggested, increase in proportion to phosphorus—thus apa-

[1] "The Data of Geochemistry," *U.S. Geol. Surv., Bull. 695* (1920).

[2] "The Composition of the Earth's Crust," *U.S. Geol. Surv. Prof. Paper 127* (1924).

[3] "Volumetric Method for Determination of Fluorine," *Indust. Eng. Chem., Anal. Ed.*, Vol. V (1933), pp. 7–10.

[4] "Volatile Constituents of Rocks: Fluorine," *Carnegie Inst. Wash. Year Book No. 34* (1934–35), pp. 98–100.

[5] "The Gases in Rocks and Some Related Problems," *Amer. Jour. Sci.*, Vol. XXXV*A* (1938), pp. 311–51; "Note on the Fluorine Content of Rocks and Ocean Bottom Samples," *ibid.*, Vol. CCXXXVIII (1940), pp. 117–28.

tite is not the chief carrier of fluorine in igneous rocks.

At the annual meeting of the American Mineralogical Society, in December, 1946, Dr. B. Wasserstein read a paper entitled "On the Geochemistry of Fluorine." From a great many data he concluded that the fluorine concentration in granite (presumably including the extensive pre-Cambrian granites and gneisses) would be 800 gm. per ton or more. This figure is twice as high as that given by Shepherd for plutonic rocks; and, since the continental granite areas exceed those of all other plutonic rocks by a factor of 20, it would seem very probable that the average concentration of fluorine in the near-surface rocks is close to 800 gm. per ton. This means that fluorine would be number 10 in amount among all elements, following titanium in the list. Even in weight percentage it would rank higher than chlorine, phosphorous, and sulphur; in terms of atom percentages it would be three times as abundant as chlorine and between two and three times as abundant as manganese.

If we accept this estimate of the fluorine content of the earth's crust, the next questions are to find out how fluorine behaves; in which reactions it takes place; in which minerals it is, or can be, present; to what extent differences of concentrations occur and why they occur. In order to answer these questions, we must find the relation between fluorine in the lithosphere and in the sea.

FLUORINE IN SEA WATER

The salt in the sea, its origin, and the manner of its delivery into the sea, have not been satisfactorily explained. Most widely accepted is the theory that the sea was originally fresh and gradually became saline through addition of salts contained in river water.

C. H. White,[6] on the other hand, believes that the primeval sea was saline; that volatile alkaline chlorides were formed prior to and above the silicates; and that they came to rest upon the primeval silicate crust and remained there until they were dissolved by the early rains and were carried as saturated brine into the growing oceans.

J. Joly[7] has held the view that the primeval atmosphere contained HCl which, mixed with rain water, was able to dissolve metals from the silicate crust and carry them as chlorides into the sea, which originally was a dilute solution of HCl without sodium. Likewise, Joly has shown that, for each kilogram of sea water, about 600 gm. of average igneous rocks would have to pass through the cycle of weathering and sedimentation in order to deliver the necessary sodium, assuming that one-third of the soda of the parent-rock had been locked up in sediments, while two-thirds still remained in the sea. Although recognizing the uncertainties involved, we shall use Joly's figure as the basis for calculating the amounts of mineral matter that have been supplied to the sea. Even if this estimate is incorrect, there should probably be no great change in the proportional relations of the figures presented in Table 1.

Table 1 demonstrates the very interesting fact that chlorine and bromine are found in sea water in such large quantities that—if the data underlying the computations are anywhere near right—a derivation from the lithosphere

[6] "Why the Sea Is Salt," *Amer. Jour. Sci.*, Vol. CCIV (1942), 714–24.

[7] "An Estimate of the Geological Age of the Earth," *Sci. Trans. Roy. Soc. Dublin*, Vol. VII (1899), pp. 23–66.

through weathering processes seems impossible. The amount of chlorine thus delivered to the sea through all geologic ages would be 290 mg. per kilogram of sea water. But 1 kg. of sea water today contains 19,300 mg., or 6,655 per cent of the chlorine thus delivered. The percentages given in the last column of Table 1 should, for each element, represent the fraction of the total amount delivered that was retained in the sea. Consequently, figures higher than 100 per cent indicate that the corresponding elements must have been present as primary constituents of the sea or else were delivered to the sea by volcanic gases and hot springs.

TABLE 1

HALOGENS PRESENT IN ROCKS AND IN SEA WATER*

	Present in Rocks (Gm/Ton)	Delivered to Each Kg. of Sea Water by Weathering Processes (Mg.)	Present in Sea (Mg. per Kg. of Water)	Percentage
F.....	800	480	1	0.2
Cl.....	480	290	19300	6655
Br.....	2	1.2	66	5500
I.....	0.3	0.18	0.05	28

* The values for Cl, Br, and I in rocks are taken from: F. W. Clarke and H. S. Washington, "The Composition of the Earth's Crust," *U.S. Geol. Surv. Prof. Paper 127* (1924); L. S. Selivanov, "Chlorine and Bromine in Massive Crystalline Rocks," *Compt. rend. (Doklady) Acad. Sci. U.R.S.S.*, Vol. XXVIII (1940), pp. 809–13; T. Fellenberg and G. Lunde, "Contribution à la géochemie de l'iode," *Norsk Geol. Tids.*, Vol. IX (1926), p. 48.

New determinations of fluorine in sea water have been made by T. G. Thompson, and H. J. Taylor, "Determination and Occurrence of Fluorides in Sea Water," *Indust. Eng. Chem., Anal. Ed.*, Vol. V (1933), pp. 87–89. See also V. M. Goldschmidt, "The Principles of the Distribution of the Chemical Elements in Minerals and Rocks," *Jour. Chem. Soc., London* (1937), pp. 655–73.

A large surplus of both chlorine and bromine is present in the sea. In sharp contrast to this, only a fraction of 1 per cent of the fluorine contained in primary rocks reaches the sea and remains in solution. By far the greatest part is returned to the lithosphere. In which rocks does it occur, and in which minerals is it concentrated? Partial answers to these questions may be obtained from the following data.

FLUORINE IN THE OSLO REGION

The region around Oslo, Norway, forms a well-known and well-defined petrographic province, containing deep-seated rocks as well as lavas and progressively metamorphosed sediments. The deep-seated rocks form a series of mildly alkaline character, ranging from monzonites through syenites and quartz-syenites to alkali-granites with a nepheline-syenite offshoot. The petrographic features of the series are in part apparent from Figure 1. A half-hundred fluorine analyses of these rocks have been worked out.[8] Of interest to this paper are the results from the igneous rocks. In Table 2 the average contents of fluorine and phosphorus pentoxide of each rock type have been listed.

Table 2 shows that the phosphorus content is not related to that of fluorine in a regular way, thus again supporting Shepherd's conclusion that apatite is not the chief carrier of fluorine. This is particularly true for the later members of the magma series—nordmarkites, ekerites, and granites, which are virtually without apatite; in them most of the fluorine is fixed in biotite and hornblende (see Fig. 2).

On the other hand, attention should be directed to three individual analyses of kauaiite (so-called "Oslo-essexite"), a coarse-grained basalt or gabbro, occurring in necks or pipes that are supposed to represent feeding channels for the extensive basalt flows of the province. In these basalts we find that the proportion

[8] T. F. W. Barth and B. Bruun, "Fluorine in the Oslo Petrographic Province," *Norske Vid. Akad. Skr. No. 8* (1945), pp. 5–12.

between fluorine and phosphorus is very nearly 1:10, indicating that all fluorine is fixed in apatite.

FLUORINE IN ROCKS AND WATERS FROM ICELAND

Determinations of fluorine and phosphorus in unaltered Icelandic basalts have been listed in Table 3. The amount of fluorine is, in each case, about one-tenth that of phosphorus, which, checking with the percentage of apatite determined in thin sections, indicates that all the fluorine is fixed in apatite. Since basalts of the Oslo region show the same relation, it would seem a natural conclusion that, generally, fluorine in basalts (and probably in gabbros) is locked up in apatite.

Two samples of dike rock from northern Iceland, strongly altered (by acid thermal waters?), have been collected by L. Hawkes and analyzed by E. S. Shepherd,[9] who found them high in fluorine; the samples gave 0.156 and 0.338 per cent fluorine.

Magmatic emanations are manifestly high in fluorine, and studies of the thermal activity in Iceland have demonstrated[10] that the acid hot springs and fumaroles are more closely related to the magmatic emanations than is the water of the alkaline hot springs, which is partly meteoric and has reacted with rocks.

From areas of acid hot springs I have only one observation: Hverarönd, east of Myvatn in northern Iceland, is an area of intense solfataric activity. The acid hot springs and fumaroles have boiled to pieces and "dissolved" basaltic rocks, eventually transforming them into clay. Basalts in various stages of alteration can be found. For analysis a basaltic lava was sampled from a crack or vent through which an intense vapor exhalation was taking place. The pores of the lava were full of incrustations, mostly of

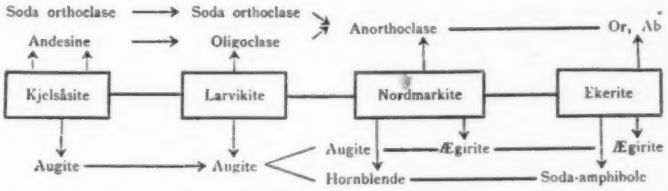

FIG. 1.—Differentiation diagram of the Oslo plutonic rocks. Lardalite is a nepheline syenite that branches off from larvikite. Akerite is a structural variety chemically related to kjelsasite.

TABLE 2

AVERAGE CONCENTRATION OF FLUORINE AND OF PHOSPHORUS IN THE PRINCIPAL TYPES OF THE OSLO IGNEOUS ROCKS

No.	Rocks	Per Cent F	Per Cent P_2O_5
1	Akerite	0.070	0.51
2	Kjelsasite	.090	.52
3	Larvikite	.078	.59
4	Lardalite	.090	.62
5	Nordmarkite	.086	.20
6	Nordm-Ekerite	.072	.08
7	Ekerite	.067	.04
8	Granite	.063	.07
9	Kauaiite	.09	.91
10	Kauaiite	.05	.55
11	Kauaiite	0.07	0.73
Weighted average		0.075	0.305

[9] "The Gases in Rocks and Some Related Problems," *Amer. Jour. Sci.*, Vol. XXXV*A* (1938), pp. 311–51.

[10] T. F. W. Barth, "Pristine and Contaminated Rock Magma and Hot-Spring Water," *Bull. volcanologique*, Ser. II, Vol. VI (1940), pp. 83–87.

sulphates (the full analysis of the rocks is given in Table 4). As seen from the analysis, this lava obviously has received fluorine delivered by the acid gases.

Alkaline hot-spring water is remarkably low in fluorine; the concentration is in the same range as that encountered in ordinary river water. Silica sinter deposited by hot springs is also low in fluorine (see Table 5). Since the hot-spring water is fed by magmatic emanations, one would expect it to contain much fluorine, unless fluorine was extracted by interaction with the contiguous rocks.

TABLE 3

UNALTERED BASALTS FROM ICELAND

Locality	Per Cent F	Per Cent P_2O_5	P_2O_5/F
Ellidaárvogur	0.002	0.02	10.0
Svinahlid	.006	.08	13.3
Hveradal	.020	.21	10.5
Ash from Vatnajökull	.022	.23	10.5
Littli Geysir	0.040	0.42	10.5
Average	0.018	0.192	10.6

In order to test this, two analyses were made.

1. A pebble of basaltic composition was found in the crater of a small, boiling spring close to Geysir. For a long time the pebble has been exposed to the boiling water (pH=8.5); it was obviously altered by the water and should have had a good chance to absorb or assimilate fluorine, if there were any. The analysis shows, however, that no fluorine was assimilated; on the contrary, the ratio P_2O_5/F indicates that some fluorine was lost.

2. A drill core of basaltic tuff was taken at 200 meters depth at Reykir in Mosfellssveit, an area of exceptional thermal activity, where the underground is soaked with hot-spring waters (pH=8.0). Again the analysis demonstrates that no fluorine was deposited in, or assimilated by, the tuff[11] (see Table 5).

The analyses of Tables 4 and 5 indicate, therefore, that at least part of the fluorine of the primary magmatic emanations is contained in the acid hot springs

TABLE 4

ALTERED LAVA EAST OF HVERARÖND

	1		2
	Weight Per Cent	Atom Per Cent	Weight Per Cent
SiO_2	42.21	49.6	10.61
TiO_2	1.87	1.6	0.47
Al_2O_3	7.69	10.6	7.84
Fe_2O_3	11.87	10.6	3.55
FeO	1.70	1.7	5.70
MnO	0.02	.....	.....
MgO	0.96	1.7	1.53
CaO	5.40	6.8	2.72
Na_2O	1.80	4.1	0.22
K_2O	0.52	0.8	0.07
H_2O^+	6.09	(45.2)	5.79
H_2O^-	5.42		24.66
CO_2	0.22	0.4	.....
P_2O_5	0.20	0.2	.....
SO_3	13.52	11.9	37.17
F	0.48	(1.8)	.....
Cl	0.02	.....	.....
Total	100.01	100.0	100.33

NOTES TO TABLE 4

None: ZrO_2, Cr_2O_3, BaO, rare earths.

1. Analysis of altered lava east of Hverarönd, northern Iceland (Barth analyst).

2. Analysis of alunogen efflorescences, Námafjall, northern Iceland (Bruun analyst).

CALCULATED MINERAL COMPOSITION OF ALTERED LAVA

Quartz and opal	31.4
Albitic feldspar	26.5
Hematite, ilmenite	12.8
Pyroxene	2.0
Calcite, apatite, fluorite	2.1
Gypsum	8.0
Alunogen*	17.2
	100.0

* Composition as stated in col. 2.

[11] A rhyolite from Yellowstone, altered by alkaline thermal waters, was tested by Fenner and found to contain no fluorine (see in E. T. Allen and A. L. Day, "Hot Springs of the Yellowstone National Park," *Carnegie Inst. Wash. Pub. No. 466* [1935], p. 471).

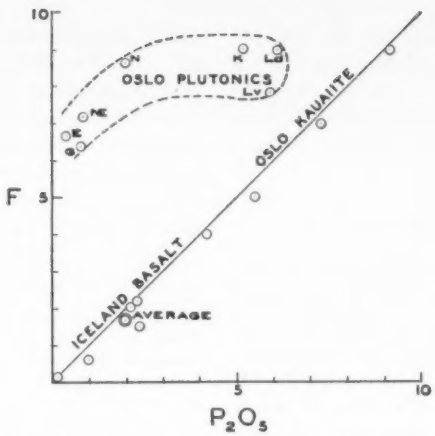

FIG. 2.—Concentration of fluorine and phosphorus in various rocks. *Ordinate:* content of F in hundredths of per cent; *abscissa:* content of P_2O_5 in tenths of 1 per cent. *E* = ekerite, *G* = granite, *K* = kjelsasite, *Ld* = lardalite, *Lv* = larvikite.

TABLE 5

CONCENTRATION OF FLUORINE IN HOT SPRINGS AND IN ROCKS ALTERED BY ALKALINE HOT-SPRING ACTION

Name of Spring	pH	F (Mg. per Liter)	Rock	Per Cent F	Per Cent P_2O_5	P_2O_5/F
Spytir	8.0	1.45	Silica sinter*	0.003	Nil	.....
Reykir in Olfus	8.0	0.55	Basaltic pebble†	.015	0.24	16
Fosshver	8.2	Nil	Drill core‡	0.006	0.06	10

* From Stekkjatunshver. † From crater of hot spring. ‡ From Reykir in Mosfellssveit.

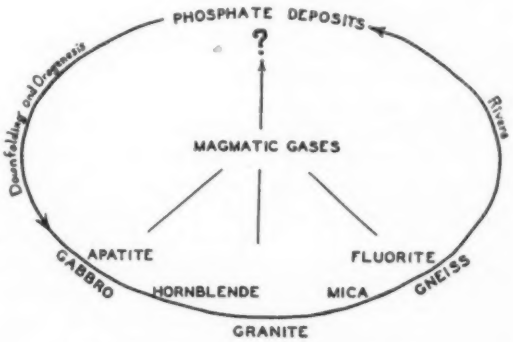

FIG. 3.—The geochemical cycle of fluorine

and fumaroles but that the alkaline hot springs have lost all primary fluorine before they emerge at the surface. But the mechanism of this extraction of fluorine is not known, nor is the location where it takes place.

CONCLUSION

It would seem that the cycle of fluorine is something like this: From inland waters and from the sea fluorine is extracted by phosphates.[12] Thereby, mineral deposits with a composition resembling fluorapatite are formed. These deposits represent a station on the road back to the igneous rocks.

The close relation of phosphorus to fluorine, so important in the sedimentary cycle, does not persist through subsequent transformations to other types of rock. In the lithosphere, fluorine and phosphorus gradually become separated as is evidenced by the fact that in the late-magmatic rocks and in the high-metamorphic sediments fluorine occurs in such minerals as mica and hornblende that contain no phosphorus.

Figure 3 illustrates graphically the geochemical cycle of fluorine.

[12] G. R. Mansfield ("The Role of Fluorine in Phosphate Deposition," *Amer. Jour. Sci.*, Vol. CCXXXVIII [1940], pp. 863–79) thinks that fluorine is caught by phosphorus compounds and locked up in insoluble deposits, whereas fluorine compounds without phosphorus generally are dissolved and carried away. Since phosphates and fluorine seem to have great affinity for each other, any conditions affecting phosphate deposition might tend also to localize the trapping of fluorine and account for its spotty occurrence in sea-bottom deposits, as noted by Shepherd.

TECTONIC FEATURES OF THE UTLEY META-RHYOLITE

O. E. GRAM

ABSTRACT

In south-central Wisconsin, outcrops of pre-Cambrian igneous rocks occur as monadnocks and ledges surrounded by Paleozoic sediments of Cambrian and Ordovician age. A careful study of such features as flow structures, chilled zones, inclusions, spheroids, gash veins, and an apparent sill leads to the conclusion that there is a minimum of eight separate flows, which were later subjected to stress in a horizontal shearing movement, which resulted in the development of an overturned anticline and syncline. During the field seasons of 1938 and 1939 the writer made a detailed study of the Utley meta-rhyolite, about 17 miles south of Berlin, Wisconsin, and a reconnaissance survey of the other igneous rocks in the area. A map of the quarry at Utley was made by a pace-and-compass survey, no other maps being available.

IGNEOUS ROCKS IN THE AREA

GENERAL RELATIONS

The Utley meta-rhyolite is one of a group of pre-Cambrian igneous rocks which occur at ten separate localities in south-central Wisconsin in an area of about 750 square miles (Fig. 1). Six of the outcrops are acidic extrusives, classed as devitrified rhyolites or meta-rhyolites; three are intrusive granites; and one is a close-to-the-surface, or hypabyssal, intrusive rhyolite. W. C. Alden[1] reports that the presence of other pre-Cambrian monadnocks beneath the Paleozoic sediments is shown by well-borings at several places in the area. In nearly all the localities the acid rocks are cut by basic dikes of altered diabase or greenstone, and in a few cases the main igneous body and the basic dikes are cut by dikes of granite porphyry.

The relation of the igneous rocks to the Paleozoic sediments is one of nonconformity. A conglomerate marks the base of the upper Cambrian St. Croix sandstone wherever it is in contact with the underlying igneous masses, the included pebbles and boulders being rhyolite fragments derived from the weathering of the pre-Cambrian rocks before Cambrian submergence. The St. Croix is the only Paleozoic sediment in direct contact with the pre-Cambrian masses.

CHEMICAL COMPOSITION AND AGE OF THE ROCKS

Chemically, the igneous rocks exposed in the area show a fairly uniform composition, and this similarity is the basis for assuming a common age. This chemical similarity extends beyond the area and into the Baraboo region, where the rhyolites have been described by J. T. Stark[2] as pre–Middle Huronian in age. Thus the Utley meta-rhyolite and the other igneous masses in the area are assumed to be pre–Middle Huronian in age.

Complete analyses of these rocks were made by W. W. Daniells, of the University of Wisconsin, and published by W. H. Hobbs and C. K. Leith.[3] Table 1 shows these analyses, with an analysis of the Baraboo rhyolite included for comparative purposes.

As shown by the table, the rocks of this area are all characterized by a high silica content. Another prominent fea-

[1] "The Quaternary Geology of Southeastern Wisconsin," *U.S. Geol. Surv. Prof. Paper 106* (1918), p. 51.

[2] "Igneous Rocks in the Baraboo District, Wisconsin," *Jour. Geol.*, Vol. XL, No. 2 (1932), pp. 119–39.

[3] "Pre-Cambrian Volcanic and Intrusive Rocks of the Fox River Valley, Wisconsin," *Univ. Wis. Bull. 158, Sci. Ser.*, Vol. III, No. 6 (1907), pp. 259–63.

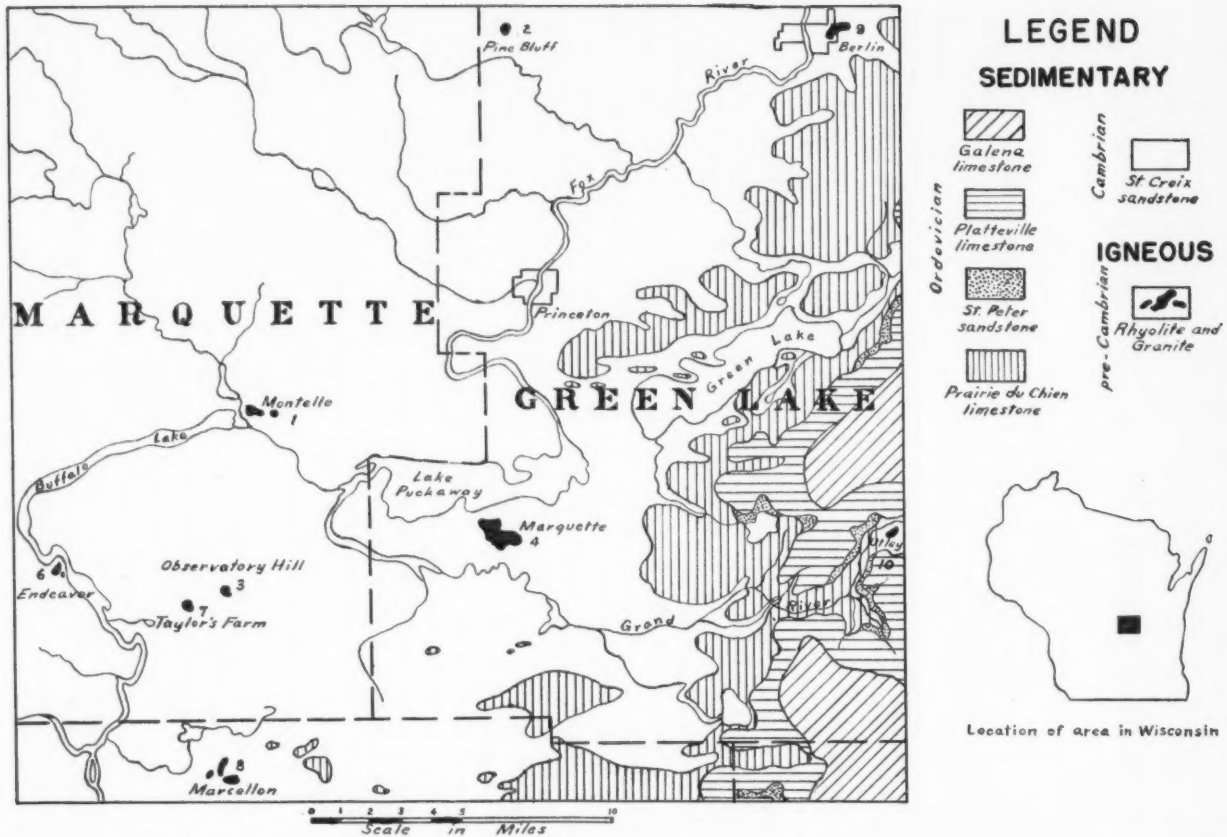

FIG. 1.—Geologic map showing location of igneous rock outcrops

ture is the predominance of soda over potash in many of the localities. In general, these chemical relationships are so striking that they strongly suggest a consanguinity of the rocks.

PREVIOUS WORK IN THE AREA

The most comprehensive work done in the area was that of Samuel Weidman[4] in 1898. He described three of the outcrops, including Utley, in detail, dealing mainly with the microscopic petrography of the rocks.

GENERAL GEOLOGY OF THE UTLEY META-RHYOLITE

PETROGRAPHIC AND CHEMICAL CHARACTER

Technically the rock at Utley should be referred to as a "porphyrytic rhyolite" in order to signify an extrusive rock as distinct from an intrusive, which, with the same petrographic character, would be termed a "rhyolite porphyry." In addition, microscopic examination plainly shows that the rhyolite has been metamorphosed, and, in conformity with the usage of the United States Geological Survey, it should be called a "meta-rhyolite." Strictly speaking, therefore, the rock at Utley is a porphyritic meta-rhyolite. For convenience in the following discussion this technical nomenclature will be disregarded, and the rock will be called a "meta-rhyolite."

TABLE 1

CHEMICAL ANALYSES OF THE IGNEOUS ROCKS

	1*	2	3	4	5	6	7	8	9	10
SiO_2	73.78	74.62	74.46	73.30	71.24	72.80	78.23	79.03	73.65	73.09
Al_2O_3	17.18	10.01	15.28	15.32	12.20	15.50	11.11	13.23	11.19	13.43
Fe_2O_3	.....	3.83	1.95	1.21	1.71	2.04	1.73	0.34	1.31	2.57
FeO	1.64	1.72	0.74	0.96	5.44	0.60	1.03	0.18	3.25	.....
CaO	0.85	2.43	0.92	1.33	0.98	0.52	0.28	0.25	2.78	2.29
MgO	.....	0.33	0.08	0.39	0.13	0.08	.....	0.07	0.51	1.03
K_2O	4.48	3.38	3.01	3.86	1.86	2.52	4.08	2.28	1.86	1.58
Na_2O	2.82	3.33	2.57	3.47	4.29	5.70	3.44	3.95	3.74	3.85
H_2O	0.12	0.24	0.58	0.26	0.81	0.43	0.25	0.19	0.44	0.72

* 1, Montello granite; 2, Pine Bluff and Waushara granites; 3, Observatory Hill rhyolite; 4, Marquette rhyolite; 5, Baraboo rhyolite; 6, Endeavor rhyolite; 7, Taylor's Farm rhyolite; 8, Marcellan rhyolite; 9, Berlin rhyolite; 10, Utley meta-rhyolite.

Another fairly detailed study of the area was made in 1906 by W. H. Hobbs and C. K. Leith.[5] They discussed the surface volcanic features in detail and cited the lithologic and chemical similarities of the rocks as proof of their consanguinity.

Since Hobbs and Leith published their report, no further detailed geologic work has been carried on in this area, although quarrying operations continuously exposed new surfaces until 1929.

The characteristic texture of the Utley meta-rhyolite is porphyritic. Quartz and feldspar phenocrysts are present in approximately equal proportions. The quartz phenocrysts possess good crystal form, have a limpid appearance, and are fairly uniform in size, averaging about $\frac{1}{16}$ inch in diameter. The feldspar phenocrysts are flesh-colored to white, are very

[4] "Pre-Cambrian Igneous Rocks of the Fox River Valley, Wisconsin," *Wis. Geol. and Nat. Hist. Surv., Bull. 3* (1898).

[5] Pp. 247–78 of ftn. 3.

FIG. 1.—Geologic map showing location of igneous rock outcrops

conspicuous by their color contrast with the dark groundmass, and show a wide variation in size, ranging from less than $\frac{1}{16}$ to $\frac{3}{8}$ inch in diameter. The groundmass is uniformly aphanitic and almost glassy in the hand specimen, although microscopic examination reveals a porphyritic texture. In color the groundmass is very dark to black, a somewhat uncommon color in rhyolites.

Quartz and feldspar are the only minerals recognizable megascopically; but the type of feldspar cannot be determined, although the flesh color is suggestive of an alkalic variety. Microscopic examination of thin sections reveals two varieties of feldspar which occur either alone or together in an interlocking structure known as "microperthite." The more common variety has the extinction angle of albite, and it is probable that the lesser variety is microcline. No orthoclase was found in any of the thin sections.

Under the microscope the groundmass of the meta-rhyolite shows a definite porphyritic texture, with quartz and feldspar making up most of the larger crystals. The presence of hornblende in the original rock is shown by altered phenocrysts with the characteristic crystal outline of hornblende. In all cases several new minerals occupy the space of the original hornblende. Samuel Weidman[6] lists eight such minerals, namely, biotite, sericite, quartz, magnetite, epidote, sphene, apatite, and zircon. The writer noted all these minerals except sphene and zircon.

The smaller crystals in the groundmass are more difficult to identify. Here Weidman lists eleven minerals, including biotite, muscovite, sericite, calcite, hornblende, sphene, epidote, apatite, zircon, magnetite, and pyrite. In addition to these minerals, the writer noted a considerable amount of chlorite, relatively small amounts of kaolinite, fairly large amounts of calcite, and two other minerals which are probably iddingsite and limonite or goethite. Fluorite is present as a secondary infiltration product, apparently having been deposited from solutions circulating along fissures. Siderite is common as a secondary filling of vugs and cavities in the spheroid zones.

[6] Pp. 30–31 of ftn. 4.

PROOF OF DEVITRIFICATION

Although the groundmass of the meta-rhyolite is holocrystalline in its present state, evidence exists to prove that this crystallization is of secondary nature and that the groundmass was originally a glass. This devitrification is in keeping with the fact that few, if any, pre-Cambrian of Paleozoic rhyolites with glassy bases have ever been reported, for all of them have been more or less completely devitrified.

The primary evidence of an initially glassy character of the groundmass is the presence of perlitic cracks, spherulites, and crystallites. According to F. F. Grout[7] and many other petrographers, these textures develop only in glasses, and their presence in a holocrystalline rock is taken as a sign of an original glassy texture.

Further evidence that the Utley meta-rhyolite is a devitrified glass is the disappearance under crossed nicols of some of the original textures of the rock. Perlitic and fluxion textures are easily recognizable in ordinary light, but these textures are lost under crossed nicols in a homogeneous holocrystalline groundmass. In the few instances in which perlitic partings do not disappear, there is pronounced crystallization along the partings. Spherulites are rare under

[7] *Petrography and Petrology* (New York: McGraw-Hill Book Co., Inc., 1932), p. 115.

crossed nicols and always tend to grade imperceptibly into the crystalline groundmass.

Devitrification also is suggested by the mottled texture common in the meta-rhyolite in which crystals of quartz and feldspar include small patches of minerals identical with the groundmass. These patches have apparently resulted from devitrification of glass which occupied embayments in quartz and feldspar crystals, a feature frequently observed in the phenocrysts of rhyolites and attributed to resorption. The mottled texture is not truly poikilitic, but because of its close resemblance might be called "pseudo-poikilitic."

The presence of spherulites is a common occurrence in a glass, as spherulites represent rapid growths in a quickly cooling magma. The relative scarcity of spherulites in the Utley meta-rhyolite suggests that they have been obliterated in the process of devitrification, and the gradation of those that are present into the holocrystalline groundmass suggests that they are partially obliterated remnants of the originals.

STRUCTURAL GEOLOGY OF THE UTLEY META-RHYOLITE

EXTRUSIVE CHARACTER OF THE ROCK

Field evidence suggesting that the Utley meta-rhyolite is an extrusive body is by no means abundant, but the few structual features present are fairly conclusive of such an origin. Flow structure is evident in one locality and is suggested in others. Chilled zones are present. Oval-shaped inclusions are distributed throughout the mass of rhyolite, and their elongation in a common direction suggests orientation in a flow. Paralleling the flow structure are several layers of spheroids, which suggest weathered surfaces of separate flows. Gash veins, developed by flows slipping over one another during folding, and volcanic breccia are other features supporting the extrusive nature of the rhyolite (Fig. 2).

The field evidence is strongly supported by laboratory evidence. In thin sections many textures may be seen which are characteristic of flows. The devitrified groundmass indicates an original glass, which in itself is suggestive of a flow. The presence of numerous crystallites, including margarites, globulites, and single and radiating trichites, gives evidence of the rapid cooling of the original magma. Fluxion or flow texture is prominent and can be observed in most of the thin sections. Generally, this texture is accentuated by the presence of numerous magnetite grains arranged in lines parallel to the direction of flow. Devitrification causes this texture to disappear under crossed nicols. Perlitic partings are common, and they are suggestive of rapid cooling, with consequent contraction in an extrusive body.

STRUCTURAL FEATURES

THE LAYERS OF SPHEROIDS

Occurring in several different zones throughout the mass of meta-rhyolite at Utley are bodies of ellipsoidal shape caused by weathering along spheroidal partings. In describing these bodies, Weidman[8] appropriately used the term "spheroid." The spheroids vary in size from $\frac{1}{4}$ to 2 or more inches in diameter. Generally, they are elongated in a northeast-southwest direction. They are composed of a core having a porphyritic texture and containing the same minerals as the normal meta-rhyolite (but with a greater proportion of kaolinite and sericite) surrounded by an alteration zone made up of kaolin, sericite, and chlorite. The kaolinite and sericite suggest weath-

[8] Pp. 6–7 of ftn. 4.

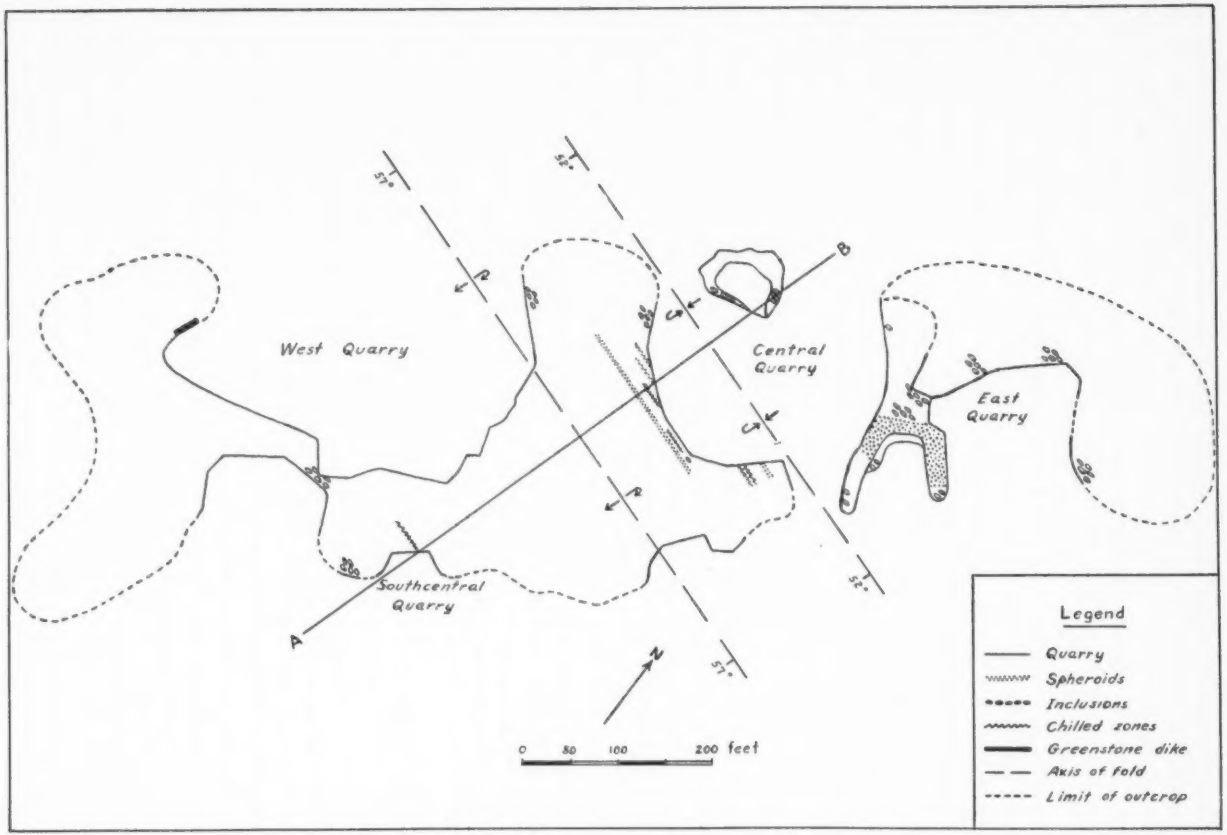

FIG. 2.—Structure map of the Utley quarry

ering of feldspars, which may have formed during devitrification along the spheroidal partings.

The spheroidal zones apparently represent tops of flows, for the most clearly defined zone shows a sharp contact with the meta-rhyolite to the north and a gradation into that to the south. If this interpretation is correct, it is probable that water solutions had easy access to the spheroid zones, with a resultant weathering of minerals along partings.

Many of the spheroids contain cavities or vugs partially or entirely filled with secondary minerals. Quartz, in well-formed crystals, is most common. Other minerals are siderite, limonite, calcite, kaolinite, and hematite. The origin of these cavities is not entirely clear, but escaping gases probably played a major role in their formation.

The spheroids occur in at least five different zones (Fig. 2), two of which probably represent the top of the same flow. One zone follows the present surface of the meta-rhyolite in the northeastern part of the outcrop. This zone is characterized by large numbers of nutlike masses or nodules on a weathered surface. The thickness of the zone is from 1 to 2 feet, but part of it may have been eroded or planed off by glacial action. Dipping 20° to the south, this zone probably represents a continuation of one of the other four spheroidal zones.

The other four layers of spheroids occur in the outcrop south and southeast of the central quarry. All dip between 80° and 85° to the south and strike N. 70° W. to N. 80° W. Their thicknesses vary from a few inches to more than 4 feet. The thickest zone occurs on a vertical face in the southeast part of the central quarry and also on the floor of the quarry west of this face. The spheroids in this zone are well defined, but more or less widely scattered, in comparison with the closely packed spheroids of the other zones. About 35 feet south of this zone is the most clearly defined layer of the outcrop, about 15 inches wide and made up of closely packed spheroids. The spheroids in this zone show a sharp contact with the meta-rhyolite to the north and a gradation into the meta-rhyolite to the south.

Between the two layers of spheroids described above is a zone on the southwest side of the central quarry which includes at least two more layers. The spheroids in these layers are small, varying between $\frac{1}{4}$ and $\frac{1}{2}$ inch in diameter, and on a weathered surface they give the appearance of rings and chains. These layers are about 4 feet apart, and both dip 82° S. and strike N. 70° W. They can be traced for only a few feet, for to the east the meta-rhyolite has been removed by quarrying, and to the west they disappear into the vertical face of the quarry wall.

Fig. 2.—Structure map of the Utley quarry

THE VOLCANIC BRECCIA

On the south face of the central quarry is a zone in which angular fragments of meta-rhyolite occur in a dark felsitic groundmass. These fragments show a small amuunt of resorption around their outer boundaries, this resorption being best shown by a slight reaction rim around the feldspar crystals and by embayments in quartz crystals. The long axes of most of these fragments parallel the strike of the spheroid layers and the other structural features. The boundaries of this zone are indistinct because of the quarrying of the meta-rhyolite to the north and the weathering of the rock to the south.

In discussing this zone, Weidman[9] implies that it is not a volcanic breccia be-

[9] *Ibid.*

cause there is no trace of vesicular texture. However, a volcanic breccia is not necessarily characterized by such a texture. Many flows fail to exhibit a vesicular texture either because there was a minimum of gas in the magma or because the viscosity of the magma was so high that the gas could not escape.

According to the definitions of various igneous breccias as given by Grout,[10] these angular fragments constitute an eruptive breccia, in contrast to flow breccia and contact breccia. In all probability the fragments represent previously solidified rhyolite in the walls of the conduit which were shattered and dispersed in a flow. This flow differs from others in the area in its extremely fine-grained felsitic character, which probably was due to a relatively high viscosity of the magma. Microscopic examination of thin sections reveals a high silica content, which undoubtedly increased the viscosity. The relatively small amount of resorption of the fragments and the concentration of the breccia in a small area are also suggestive of a magma of high viscosity, for in a more fluid magma there would be expected more resorption of the included fragments, as well as a wider dispersion of them throughout the flow.

FLOW STRUCTURE

A few feet northwest of the breccia is a limited zone in which excellent megascopic flow structure is developed. Striking N. 85° W. and dipping 82° S., this structure is clearly defined by weathering, which causes small ridges to stand out on the surface, where the crystals have a linear arrangement. The strike reading cannot be considered accurate because of the very limited exposure of the structure.

[10] P. 119 of ftn. 7.

INCLUSIONS

Widely scattered throughout all parts of the outcrop are many incusions which are ellipsoidal masses of meta-rhyolite, differing from the normal rock in the greater size of the quartz and feldspar phenocrysts and in the extreme darkness of the groundmass. They vary in size from 1 to 8 inches in their long diameter, and most of them are elongated in an east-northeast direction. Many occur singly, but others are found in groups (Fig. 2.).

Dip readings were obtained in only a few instances, for most of the inclusions are seen on horizontal surfaces. Single inclusions on vertical faces of the east quarry vary in dip from 18° to 22° S., with the exception of one, which dips 7° S. On the northeast wall of the pit in the central quarry, ten inclusions in two groups about 12 feet apart occur on a vertical face, and all of them dip 22° S. An interesting characteristic is the flatness of these inclusions in the plane of dip and strike, all being three to six times as long in the direction of dip as they are normal to it. This suggests flattening or deformation through folding.

The origin of the inclusions is not entirely clear. They could have originated in at least three different ways, namely, (1) fragmental material blown into the air in a semiplastic state and falling back into the flows, (2) erosional fragments of solidified flows being engulfed in fresh flows, and (3) solidified rhyolite, which was torn loose from the walls of the conduit by rising magma, which flowed out onto the surface. The ellipsoidal shape of the inclusions can be explained by any of the three theories; but the scattered distribution, the large size of the phenocrysts, and the character of the groundmass are most satisfactorily explained by the third theory. If this hypothesis is

correct, the inclusions were formed in the same manner as the eruptive breccia; but, because they were included in a flow of lower viscosity, their angularity was destroyed by resorption, and they assumed an ellipsoidal shape oriented in the direction of the flow. The increased size of the phenocrysts is probably due to renewed crystal growth when the fragments were included within the hot magma, and the increased size of grain in the groundmass is similarly explained. Reaction rims around phenocrysts near the contact between the inclusions and the meta-rhyolite are common, as would be expected under this theory.

GASH VEINS

On the east face of the central quarry is a series of small quartz stringers and lenses, which appear to be gash veins, filling fractures which presumably developed during folding of the flows. In one zone the veins run out horizontally to the south from two joint planes, which dip 18° S. In the other zone, a few feet to the north, the veins run out horizontally on both sides of a joint plane. A theoretical application of horizontal compression and vertical tension to these zones suggests that the rhyolite above each joint moved north with respect to the meta-rhyolite below, this conclusion being in harmony with the structure of the outcrop.

CHILLED ZONES

Near the top of a vertical face in the south-central quarry is a zone of dark aphanitic rock about ½ inch thick, striking N. 80° W. and dipping 30° S. Presumably representing the chilled surface of a flow, it is the only clue to the structure of the southern part of the outcrop.

In the central part of the outcrop, on the southeast side of the central quarry, are two well-defined chilled zones, which represent the top and bottom of a flow, a sill, or the edges of a dike. The relations of this body to the surrounding meta-rhyolite are confusing. The contact between the chilled zones to the south and the meta-rhyolite is very irregular, being fairly smooth near the base of the quarry, quite wavy halfway to the top, and extremely jagged near the top. The other chilled zone is more regular. Between these two zones the rock grades into a meta-rhyolite that is almost identical with the main mass of meta-rhyolite. In a few places the chilled zone disappears, and the porphyritic meta-rhyolite runs out to the contact, the latter, however, remaining distinct. In the meta-rhyolite on both sides of this body is a well-defined banding which is due to an alignment of feldspar phenocrysts parallel to the contact. This banding is noticeable only in the lower part of the zone.

The strike of this body cannot be determined with any degree of accuracy because of its very limited exposure at the top of the quarry, but the small zone exposed strikes approximately N. 65° W. and dips 82° S. to vertical. In thickness it ranges from 2 feet near the base of the quarry to almost 5 feet at the top. Mineralogically its composition is identical with the main mass of meta-rhyolite.

No conclusive evidence exists to prove whether this body is a flow, a dike, or a sill, but the writer favors a sill. The fact that the banding of feldspar phenocrysts occurs on both sides of the body suggests that it is an intrusion rather than a flow, for exomorphic effects of a flow would be limited to the rhyolite beneath the flow. As previously stated, the coarse grained phase of this body in places extends all the way to the contact, thus eliminating the chilled zone. This is an improbable occurrence in a flow but is not uncommon in an intrusive body. Further evidence

favoring an intrusion is negative and includes a lack of fluxion texture in thin sections, a lack of xenoliths, and a lack of vesicular texture.

INTERPRETATION OF THE STRUCTURE

From the foregoing evidence there seems little doubt that the Utley meta-rhyolite is a series of extrusions. Eight separate flows are recognized, although in some cases the evidence is meager. Spheroids mark the upper surface of at least four, and possibly five, flows. The sill-like body with its chilled zones presumably separates two flows, for if it is a sill it would be expected to intrude between flows. The chilled zone in the south-central part of the outcrop marks the top of another flow. The volcanic breccia may or may not mark a separate flow, and the spheroid zone between the central and east quarries may represent the surface of still another flow unless it is a continuation of one of the spheroid layers to the southwest. The thickness of the definitely established flows ranges from 4 to 40 feet or more.

The attitude of the various features described in the preceding pages gives a definite clue to the geologic structure of the meta-rhyolite outcrop. All features with sufficient exposure to permit accurate strike readings to be taken show a strike ranging from N. 70° W. to N. 80° W. and the remaining features with limited exposures closely approximate this strike. The dip readings obtained are strongly indicative of an overturned anticline (Fig. 3) with a south flank dipping 30° S., as shown by the chilled zone in the south-central quarry, and a north flank overturned to 82° S., as shown by the sill (?), chilled zones, and spheroids on the south side of the central quarry. The spheroids and inclusions in the northern part of the outcrop show a reversal, forming a syncline with a north flank dipping approximately 22° S. The axes of the folds strike N. 75° W., and the axial plane of the overturned anticline dips 57° S., while that of the syncline dips 52° S. (Fig. 2).

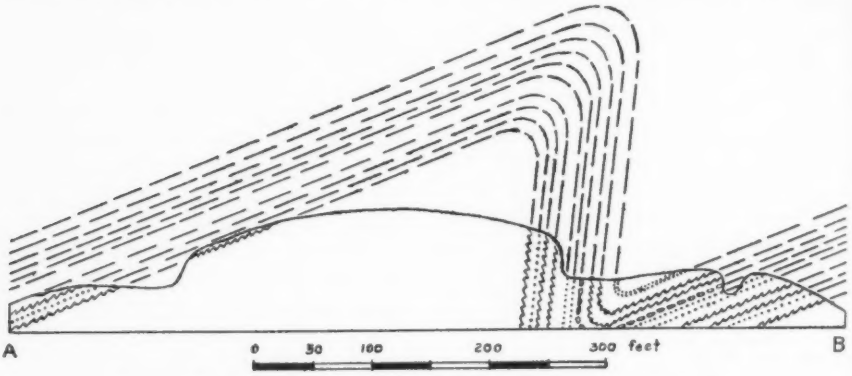

FIG. 3.—Theoretical structure section along line A–B. (See Fig. 2 for legend)

THE JOINT SYSTEM

In an attempt to determine the type of regional stress which caused the deformation of the rhyolite flows at Utley, the writer spent several days mapping the major joints or master-joints. After eliminating from consideration those joints

which were apparently related to the joints in the associated sediments in the area, three prominent sets were discovered, these sets striking N. 70° E., N. 2° E., and N. 32° W. Other joints were grouped into less prominent sets, the most prominent of which had a strike of N. 70° W., which parallels the strike of the folds, thus suggesting that they are tension joints. This conclusion is further justified by the very steep dip of these joints, their curved and irregular surfaces, and the fact that their occurrence is limited mainly to the area which roughly corresponds to the crest of the anticline.

Both the steeply dipping N. 32° W. and the N. 70° E. joints show characteristics of true shear joints in their smooth, plane surfaces. Nearly horizontal slickensides are common, especially along the planes of the N. 70° E. set. If these are true shear joints, they should furnish a clue to the direction of regional stresses.

Resistance to shearing along fracture planes is a composite of the strength of the rocks against shearing and their frictional resistance to motion.[11] The rupture has been proved theoretically to be 45° with the direction of stress, provided that friction is neglected. Actually, the effect of friction will cause this angle to approach 30°–35° with the direction of compressive stress in brittle material. According to the shear theory, the direction of maximum compressive stress will always bisect the acute angle between planes of fracture, provided that the stress is nonrotational. This theory cannot be applied to the Utley meta-rhyolite because the direction of maximum stress would have had to be from N. 71° W., in order to bisect the acute angle between the major shear planes. This direction parallels the axes of folding and is therefore impossible. G. F. Becker,[12] however, suggested that fracturing occurs where the strain is greatest. In other words, the plane of maximum strain will be a plane of fracture, and the obtuse angle between sets of fractures will face the direction of nonrotational stress.[13] By applying this theory, known as the "strain theory," to the joints and folds at Utley, the direction of maximum compression would have been from N. 19° E. A nonrotational stress from such a direction would develop the existing joint pattern and the folds at Utley almost perfectly, for the stress would be at right angles to the strike of the folds, would bisect the obtuse angle between the shear joints, and would produce the N. 70° W. tension joints. The N. 2° E. joints would not be entirely in accordance with this pattern, but friction could cause the discrepancy from the theoretically perfect development. However, an application of the strain ellipsoid to this pattern shows that under a nonrotational stress of this type it would be necessary to have the easiest relief lateral instead of vertical, in order to obtain the joint pattern existing at Utley. Because it is difficult to reconcile easy lateral relief with folding of any great extent and because compressive stresses are more often applied tangentially with a resulting rotational strain, a horizontal nonrotational stress such as described above must be considered to be in a doubtful category.

If a rotational stress in the form of a horizontal shearing movement is applied to the Utley rhyolite, the existing joint

[11] C. M. Nevin, *Principles of Structural Geology* (New York: John Wiley & Sons, Inc., 1936), p. 17.

[12] "Finite Homogeneous Strain, Flow, and Rupture of Rocks," *Bull. Geol. Soc. Amer.*, Vol. IV (1893), pp. 13–90.

[13] Recent laboratory experiments by Griggs have tended to discredit Becker's analysis.

pattern and the folds can be explained. Experimental work by W. J. Mead[14] showed that horizontal shearing movements first produced tension cracks inclined about 45° to the direction of movement. These fractures were immediately followed by two sets of nearly vertical shear fractures, one striking parallel to the direction of movement and the other parallel to the free side. Folding in such a case developed at approximately right angles to the first tension fractures. If the deformation at Utley was due to a horizontal shearing couple from N. 32° W. and S. 32° E., the first fractures to form would tend approximately N. 13° E. Since friction would cause a variation in the inclination of these fractures to the direction of shearing movement, the N. 2° E. joints might easily be the primary tension fractures. The two sets of shear fractures would then correspond to the N. 70° E. joints, which closely parallel the free side, and the N. 32° W. joints, which parallel the direction of movement. At the same time, folding would occur, and the axes of the folds would trend N. 77° W., which closely corresponds to the trend of the folds at Utley and which would give rise to tension fractures, such as those striking N. 70° W. However, the folds developed in Mead's experiments are difficult to explain because of the N. 71° W. easy lateral relief developed under a rotational stress of this type.

The writer feels that no definite conclusions regarding stress conditions in the area can be drawn until such time as all the other igneous outcrops are studied in detail. The joint system at Berlin, about 17 miles north of Utley, is extremely complicated and does not fit the pattern at Utley. However, the Berlin rhyolite offers one clue in its very pronounced cleavage system, this cleavage paralleling the northeast axis of the outcrop. Since cleavage develops in a plane normal to the shortening, it is probable that the stress at Berlin operated from a northwesternly or southeasterly direction. This fact suggests that a horizontal shearing movement from N. 32° W to S. 32° E. at Utley must be considered as a strong possibility.

ACKNOWLEDGMENTS.—The writer is especially indebted to Dr. J. T. Stark, of Northwestern University, for his assistance in the field and laboratory and for his valuable suggestions and criticisms; to Dr. A. L. Howland, who supplied several valuable suggestions pertaining to the petrographic character of the rhyolite and who obligingly checked the writer's microscopic identification of minerals in thin sections of the rhyolite; and to Dr. R. M. Garrels for his field assistance as well as his stimulating suggestions pertaining to some of the theoretical data. The writer also acknowledges the co-operation of the Wisconsin Granite Company, which granted permission to work in the quarry.

[14] "Notes on the Mechanics of Geologic Structure," *Jour. Geol.* Vol. XXVIII (1920), pp. 505–23.

DISCOVERY OF LETTERS BY LYELL AND DARWIN

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ABSTRACT

Original letters written by Sir Charles Lyell to the Rev. Charles Kingsley and the Rev. John Pye Smith and also a letter from Charles Darwin to James Croll on alternate glaciation of northern and southern hemispheres were found recently between the pages of the first edition (Vol. III) of Lyell's *Principles of Geology*. The Lyell letters do not appear in previously published biographical material on Lyell. In his letter to Kingsley, Lyell expresses his interest in the exploration of the Green and Colorado rivers by John Wesley Powell. To Dr. Smith he reveals his belief that the waves of immigration of Europeans into the United States in the 1840's, "like the irruptions of the Goths & Vandals, are doing their utmost to overwhelm the indigenous civilization, but against which a noble stand is making."

While reading Volume III of the first edition of the *Principles of Geology* by Charles Lyell[1] recently, the writer was agreeably surprised to discover two original letters written by Lyell and, on further examination of the book, a letter sent by Charles Darwin to James Croll on November 24, 1868, on the subject of alternate glaciation in the northern and southern hemispheres. The letters, loosely placed between the pages of the *Principles*, are in excellent condition. The Detroit bookseller from whom the writer purchased the book, with the companion volumes, was unable to provide any clue to their previous ownership, and the writer has no pertinent information which would permit tracing the letters along their evidently devious travels.

The Lyell letters are to the Rev. Charles Kingsley, written April 26, 1871, and to the Rev. John Pye Smith (June 2, 1849). From an examination of the known Lyell correspondence, these appear to be heretofore unpublished letters. The letter from Darwin to Croll evidently was written by Mrs. Darwin but is signed by Darwin and is mentioned in the two-volume compilation, *More Letters of Charles Darwin*.[2]

Few records of Lyell's activity during the six years prior to his death in 1875 are preserved, but these few show that the great uniformitarian retained his keen interest in geology to the end and showed no evidence of declining mental strength.

The note to Kingsley is on stationery bearing the Lyell address, 73 Harley Street, London W, embossed in blue, and was written four years before Lyell's death, as he was revising his tenth edition of the *Principles*. The letter reads as follows:

MY DEAR KINGSLEY

On my return from a geological Easter tour to Devon and Cornwall, I find your very kind note & very curious & instructive (?) Report by Major Powell on the cañons of the Rio Colorado. I must read it again when the work is out as I cannot yet fully understand the country. The *chines* as they call them in Hants near Xchurch & Bournemouth are I suppose diminutive cañons.

I hope soon to hear an account of yourself and family.

My wife desires her kindest remembrances & believe me

Ever most truly ys

CHA LYELL

"The 'chines' of Hants (Hampshire) and the Isle of Wight are, in the words of Horace B. Woodward, "gullies which

[1] *Principles of Geology* (3 vols.; London: John Murray, 1830–33).

[2] *More Letters of Charles Darwin*, ed. Francis Darwin and A. C. Seward (London: John Murray, 1903).

73. Harley St
London. W.

April 6 1871

My dear Kingsley

On my return from a visit to the East as far as Truro in Cornwall I found your very kind note & very curious & instructive Report by Major Powell on the canons of the Rio Colorado. I must read it again that the work is not as I cannot yet fully understand the country. The chiefs as they call them in Utah were Mr Brigham Young & Heber C. Kimball. Powell's description of the latter as a "huge sensualist" seems to be borne out by the accounts of Mormonism which I have read. I am very glad to find that you are desirous to understand the Mormon question & believe me

Yours very truly

Chas Lyell

have been formed by the action of springs in making their way over the cliffs into the sea."[3]

Lyell's excursions frequently took him to the coast in the Christchurch-Bournemouth area, where the Eocene strata have been deeply incised.

When the letter was written, Major Powell was at Green River, Wyoming, preparing for his second trip down the Green and the Colorado and had probably already made his observation that "the Uintas rose no faster than the Green could erode."

The energetic and versatile Powell, who had lost part of an arm at Shiloh and had collected fossils while his men dug trenches before Vicksburg, now was destined to write one of the most dramatic chapters in American geology. He could not have known that the vast canyons of the Southwest were being compared across the Atlantic to "the chines in Hants" or that Lyell, the master whose ideas Powell had championed in America in the face of stubborn opposition, was eagerly awaiting publication of Powell's papers, "the better to understand the country."

The four tours of Lyell in the United States three decades earlier had taken him only to the Mississippi. We can only wonder what impetus he might have given to the development of American geology, had circumstances permitted him to travel into the then still untamed West.

Kingsley, canon of Chester, sociologist, and geologist, is better known as the author of *Westward Ho!* and *The Water Babies*. He also wrote *Town Geology*, an introduction to geology for the people of Chester. He met Darwin in 1854, at the Linnaean Society and recalled Darwin as "a quiet, meek man, very anxious to know whether I really 'denied the atonement,' on which point, I think, I satisfied him."

Lyell met Kingsley in 1860, the year in which the exuberant Huxley, slapping the knees of a friend next to him and uttering, "The Lord hath delivered him into my hands," launched into his reply to the Bishop of Oxford, who was at the time attacking Darwin's newly published *Origin of Species*.

Darwin, Lyell, and Kingsley kept up a sprightly correspondence through the years, and the influence of the great naturalists on the theologian-geologist is apparent in Kingsley's letter to a friend, the Rev. F. D. Maurice, in 1863:

> I am very busy working out points of Natural Theology, by the strange light of Huxley, Darwin and Lyell. The state of the scientific mind is most curious; Darwin is conquering everywhere, and rushing in like a flood, by the mere force of truth and fact. The one or two who hold out are forced to try all sorts of subterfuges as to fact, or else by evoking the *odium theologicum*.

The four visits of Lyell to the United States were made during a great tide of immigration, such as the country had never seen before. In 1844 alone, 297,000 European emigrants came to America. In 1845, during his second visit, 114,000 were admitted. The Irish famine brought another deluge of newcomers in the succeeding years, and by 1850 the total population was in excess of 23,000,000.

What he saw in Boston, in New York, and in the rude hinterland and the babble of foreign tongues that he heard caused Lyell much concern, as witness his letter to Dr. Pye Smith in 1849, following his return to England:

> My dear Sir
>
> I have given orders to my publisher to send a copy of my book to Jackson & Wolfe 18 St. Paul's Ch y'd, of which I beg your acceptance. It is entitled a Second Visit to the United States & I hope that none of my allusions in the 11th

[3] *Geology of England and Wales* (London: Longmans, Green & Co., 1876).

and 12 Chapters v. 1. to the Congregational or Independent Churches are offensive to your views in reference to the bearing of the organization of such churches on the educational movement now in progress in those parts of the Union which are least kept back by emigration from Europe which like the irruptions of the Goths & Vandals are doing their utmost to overwhelm the indigenous civilization but against which a noble stand is making.

Believe me
Sincerely yours
CHA LYELL

Dr. Pye Smith

The Rev. John Pye Smith (1774–1851) was pastor for forty-three years of the (Independent) Gravel Pits Chapel at Homerton, a Fellow of the Royal Society, and author of *The Mosaic Account of the Creation and the Deluge Illustrated by the Discoveries of Modern Science* (1837) and *Scripture and Geology* (1839).

The library of Dr. Smith no doubt already held a copy of Lyell's *Travels in North America, with Geological Observations on the United States, Canada and Nova Scotia*,[4] the nongeologic chapters therein containing this paragraph:

The state of Ohio has always punctually discharged the interest of her debt by direct taxes imposed for that special purpose, although there has been a deficit from the beginning on the proceeds of her public works. She is of recent origin, and her growth has been more rank and luxuriant than that of any other State of the Union. An influx of illiterate Irish, Welsh and Westphalian settlers, has tended to lower the educational qualifications of her electors, considered as a whole; but she came of a good New-England stock, which, like the philosopher's stone, has converted much of baser metal into gold.

Now, four years after the appearance of this volume, Dr. Pye Smith, upon receiving his complimentary copy of *Second Visit to the United States*, found additional fears concerning the future of the Union in Lyell's description of a "repeal meeting" in Boston, where "an orator with an Irish accent, addressed the crowd on the sufferings of the Irish people precisely as if he had forgotten on which side of the Atlantic he then was!"

The Darwin letter of 1868 to James Croll, one of the documents found between the pages of my copy of Lyell's *Principles*, is as follows:

DEAR SIR

I have read with the greatest interest the last paper which you have kindly sent me. If we are to admit that all the scored rocks throughout the more level parts of the United States result from true glacier action it is a most wonderful conclusion & you certainly make out a very strong case; so I suppose I must give up one more cherished belief. But my object in writing is to trespass on your kindness & ask a question, which I dare say I c'd answer for myself by reading more carefully, as I hope hereafter to do, all your papers; but I shall feel much more confidence in a brief reply from you.

Am I right in supposing that you believe that the glacial periods have always occurred alternately in the northern and southern hemispheres; so that the erratic deposits which I have described in the S. parts of America & the glacial work in New Zealand c'd not have been simultaneous with our glacial period. From the glacial deposits occurring all round the Northern hemisphere & from such deposits appearing in S. America to be as recent as in the North, & lastly, from there being some evidence of the lower descent of glaciers all along the Cordillera, I inferred that the whole world was at this period cooler. It did not appear to me justifiable without distinct evidence to suppose that the N. and S. glacial deposits belonged to distinct epochs; tho' it wd have been an immense relief to my mind if I cd have assumed that this had been the case.

Secondly, do you believe that during the glacial period in one hemisphere, the opposite hemisphere actually becomes warmer, or does it merely retain the same temperature as before?

I do not ask these questions out of mere curiosity, but I have to prepare a new ed. of my Origin of Species, & am anxious to say a few words on this subject on yr authority. I hope you will excuse my troubling you.

Pray believe me
Very faithfully yours
(*Sgd.*) CHARLES DARWIN

Darwin was seeking an explanation for the observed distribution of plants

[4] 2 vols.; London: John Murray, 1845.

11 Harley St
June 2d 1839

My dear Sir

I have given orders to my publisher to send a copy of my book & Jackson & Walford's 6/ Lectures the 5th of which I hope you accept[ed] - I visited a second time & the hundred Sheep & 9 Lepers that were of my other cases in the 11th & 12 chapters of the congregational & Independent Churches are opposed to your view in reference to the bearing of the congregation of such churches on

the education & movements in progress in those parts of the Union which are least affected by emigration from Europe which like the exemptions of the Islands are doing those almost the only obstacles the Indigenous civilization has against which a noble stand is making. Believe me

Yours truly

Wm Ellery Channing

J. Pye Smith

I do not ask these questions out of mere curiosity; but I have to prepare a new ed. of my Origin of species, & am anxious to say a few words on this subject on yr authority: I hope that you will excuse my troubling you.

Pray believe me
Very faithfully yours

Charles Darwin

Down
Bromley
Kent. S.E.
Nov 24 1868

Dear Sir

I have read with the greatest interest the last paper which you have kindly sent me. If we are to admit that all the scored rocks throughout the more level parts of the United States result from true glacier action it is a most wonderful conclusion, & you certainly make out a very strong case; so I suppose I must give up one more cherished belief. But my object in writing is to trespass on your kindness & ask a question, which I dare say I'd answer for myself by reading more carefully, as I hope hereafter to do, all your papers; but I shall feel much more confidence in a brief reply from you. Am I right in supposing that you believe that the glacial periods have always occurred alternately in the northern & southern hemispheres; so that the glacial erratic deposits which I have described in the S. parts of America, & the glacial work in New Zealand did not have been simultaneous with our glacial period. From the glacial deposits occurring all round the Northern hemisphere, & from such deposits appearing in S. America to be as recent as in the North, & lastly, from there being some evidence of the former lower descent of glaciers all along the Cordilleras, I inferred that the whole world was at this period cooler. It did not appear to me justifiable without distinct evidence to suppose that the N. & S glacial deposits belonged to distinct epochs, tho' it would have been an immense relief to my mind if I'd have assumed that this had been the case. Secondly, do you believe that during the glacial period in one hemisphere, the opposite hemisphere is actually rendered warmer, or does it merely retain the same temperature as before?

and animals and was turning more and more to the astronomical theory of Croll (1821–90), the Scottish geologist and climatologist, and away from the opinion of his old friend, Lyell, who consistently maintained that geographical changes are the principal, and not the subsidiary, causes of glaciation.

A zealous disciple of Lyell ever since he had first read Lyell's *Principles* as a youth while on the epochal cruise of the "Beagle," Darwin now chose to differ from Lyell regarding the validity of Croll's views. In 1866, two years before the letter to Croll was written, Lyell wrote to Darwin as follows:

> I have been doing my best to do justice to the astronomical causes of former changes of climate, as I think you will see in my new edition [the tenth, first vol. published in November, 1866, the second in 1868] but I am more than ever convinced that the geographical changes are, as I have always maintained, the principal and not the subsidiary ones. . . . . In my winter of the great year, I gave you in 1830 cold enough to annihilate every living thing. The ice now prevailing at both poles is owing to an abnormal excess of land, as I shall show by calculation. Variations in eccentricity [of the orbit] have no doubt intensified the cold when certain geographical combinations favored them, but only in exceptional cases, such as ought to have occurred very rarely, as paleontology proves to be the case.
>
> Ever most truly yours,
>
> CHARLES LYELL

For Darwin, however, the Croll theory gained in appeal, and his letter to Croll followed. This letter was followed by another from Darwin, a few months later, in which Darwin wrote Croll:

> Sir C. Lyell, who is staying here, is very unwilling to admit the greater warmth of the S. hemisphere during the Glacial period in the N; but, as I have told him, this conclusion which you have arrived at from physical considerations, explains so well whole classes of facts in distribution, that I must joyfully accept it; indeed, I go so far as to think that your conclusion is strengthened by the facts of distribution. . . . .

In 1872, Darwin, in his sixth edition of the *Origin of Species*, abandoned his cautiously worded statement of earlier editions, in which he indicated belief in a world entirely colder during glacial times. He now gave considerable space to Croll's ideas, adding that "this [Croll's] conclusion throws so much light on geographical distribution [of animals and plants] that I am strongly inclined to trust it. . . . ."

In the same year there appeared Lyell's eleventh edition of the *Principles of Geology*, revised, and Lyell stands firm, stating:

> Mr. Darwin in the last edition of his Origin of Species has inclined toward adopting Mr. Croll's theory of alternate glaciation and perpetual spring in the opposite hemispheres, on the ground that it would account for some of the anomalies in the distribution of animals and plants, by affording a refuge for tropical life during a period of extreme cold.
>
> But . . . . until we know what climate the countries now inhabited by tropical animals and plants were enjoying in glacial times it is premature to contend with imaginary difficulties as to the survival of forms which would have been extinguished if the snow and ice had been universal down to Lat. 55, even over one hemisphere at a time.

Today, after seventy-five years of research and speculation, Lyell's view of the problem still agrees with that of leading students of the Pleistocene, a tribute to the sagacity of the man to whom Darwin dedicated the second edition of his journal[5] of the voyage of "H.M.S. Beagle" with his acknowledgment that "the chief part of whatever scientific merit this journal and the other works of the author may possess, has been derived from studying the well-known and admirable *Principles of Geology*."

[5] Charles Darwin: *Journal of Researches into the Natural History and Geology of the Countries Visited during the Voyage of H.M.S. Beagle Round the World* (London: John Murray, 1852).

REVIEWS

The Pleistocene Period: Its Climate, Chronology and Faunal Successions. By FREDERICK E. ZEUNER. London: Ray Society; sold by Bernard Quaritch, Ltd., 1945. Pp. xii+322; figs. 76. 46*s*.

This book is "entirely concerned with the evidence for the relative and absolute chronology of the Pleistocene, for the various types of environment existing at certain times in certain areas, and with the evolution of the fauna in deposits of known age" (p. iii). Data on climatic and geographic changes and on stratigraphy are reviewed and marshaled into a relative, stratigraphic chronology of the Ice Age; the modern astronomic hypothesis of the Pleistocene glacials and interglacials is reviewed; and the relative and the absolute chronologies are merged. The evolution of the modern mammals receives special attention. The treatment is essentially confined to the Old World.

An introductory chapter is devoted to some principles of Pleistocene stratigraphy (pp. 1–29). The subdivisions of this age are primarily based on changes in climate (p. 2); and the evidence is supplied by till, moraines, and other glacial deposits, loess, frost phenomena, soils due to chemical weathering, river terraces, fauna, and flora. Because an advancing ice normally destroys older glacial deposits, the formations of the periglacial belt furnish more complete records (p. 28). These foundations of the relative chronology are lucidly though briefly discussed.

Chapters ii–iv deal with the relative chronology proper (pp. 30–135). In northern Germany three separate glaciations are distinguished, viz., the Elster, Saale, and the Last Glacial, corresponding to the Mindel, Riss, and Würm (p. 38). The Last Glacial probably included the Warthe, Weichsel, and Pomeranian stages. However, the age of the Warthe, which is comparable to the Iowan of North America, remains an unsolved problem (pp. 33, 35, 39). In the Alps there may have been four main glaciations, as Penck and Brückner established in 1909, using glaciofluvial gravels: the Günz, Mindel, Riss, and Würm (p. 41). Late studies are believed to prove that the Würm comprised three maxima; each of the others, two maxima (pp. 47, 48); but this is not conceded by Albrecht Penck.[1] Besides, there appear to have been several pre-Günz glacial ages. An attempt at correlating the major Pleistocene climatic ages in Scandinavia-Germany, the Alps, and North America is presented on page 53.

The periglacial zone is the belt of frost climate surrounding an ice sheet (p. 55). While the modern periglacial zone coincides with the tundra, that of the glacials included also a peripheral loess steppe belt which graded into the forest zone. The loess steppe was dry and cold. Its subsoil was permanently frozen, and its highest monthly mean was probably below 10° C. (50° F.). The periglacial area, which was temperate during interglacials and interstadials, has furnished the most complete record of the climatic variations during the Pleistocene (p. 56). Climatic fluctuations are recorded by river terraces within the zone (pp. 25, 56). During the oncoming of a glacial the forests withdrew from high summits and ridges and frost action increased, supplying streams with an abundance of debris, which they carried downvalley. When the glaciation waned, the vegetation grew denser and returned to higher levels, and the streams began to sink their beds in the deposits of the preceding cold age, converting them into terraces. The river terraces reveal at least eight cold ages, grouped in four pairs (p. 63). Climatic changes are also recorded by a succession of loess beds (cold and dry climate) and weathering soils (temperate and humid climate) (p. 64). In Germany there are at least two young loesses of the Last Glacial and three old loesses (p. 73). A correlation of periglacial river terraces and loess beds with glacial stages is suggested on page 79.

The climatic chronology of northern France, based on loess and solifluction recording the glacials and on weathering indicating the interglacials and interstadials, has been correlated with low (glacial) and high (interglacial) sea-levels, respectively, which controlled the terraces of the Somme River (pp. 92, 99).

To sum up: In continental Europe the Last

[1] "Die Strahlungstheorie und die geologische Zeitrechnung," *Zeitschr. d. Gessellsch. f. Erdkunde zu Berlin*, Jahrgang 1938, pp. 321–50; see p. 336.

Glacial comprised two main stages and regionally a third stage. The two intermediate glacials each had two stages, and the earliest glacial two stages in the Alps, one stage elsewhere. The interglacials were mild (p. 99). This climatic succession, based purely on geological observations, is called the detailed relative chronology (p. 100).

Geographical separation, oceanic climate, and several centers of glaciation make it difficult to establish a Pleistocene climatic chronology for the British Isles (p. 101). However, the till deposits seem to indicate three cold ages of the Last Glacial, two extensive glaciations which probably were correlatives of the Saale and the Elster, and two minor cold ages in Crag times which probably corresponded to the Günz (p. 114). In the Thames basin buried channels, fillings, and abandoned terraces record glacials, interglacials, and interstadials which seem to correspond to similar stages on the Continent (pp. 133, 135).

Chapter v, pages 136–65, is entitled "The Astronomincal Theory as the Basis of an Absolute Chronology of the Pleistocene." The astronomical elements considered are the inclination of the earth's axis (the obliquity of the ecliptic; mean length of periodicity forty thousand years), the eccentricity of the earth's orbit (mean length of periodicity ninety-two thousand years), and the longitude of the perihelion (precession of the equinoxes; mean length of periodicity twenty-one thousand years). Formulas for calculating the secular changes of these elements were developed by the American astronomic mathematician J. N. Stockwell in 1873, and the calculations were made by the German, Ludwig Pilgrim, in 1904. From Pilgrim's determinations the Serbian mathematician and physicist Milutin Milankovitch in 1920 deduced the consequent secular fluctuations of the solar radiation received at the outer limit of the earth's atmosphere; and in 1924, at the request of the German climatologist Wladimir Köppen, he expressed the radiation changes on the latitudes 55°, 60°, and 65° N. during the summer half-year of the last six hundred and fifty thousand years in the form of latitudinal equivalents. Köppen interpreted Milankovitch's radiation graph as a graph of the Pleistocene glacials and interglacials, and he correlated the glacials with the minima of summer radiation and the associated maxima of winter insolation.

Later the Serbian astronomer V. Michkovitch, using the formulas of 1843 of the French mathematician U. J. J. Leverrier, duplicated Pilgrim's calculations of the secular changes of the mentioned astronomical elements; and Milankovitch again calculated the corresponding radiation changes and constructed a graph of the summerly variations on latitude 65° N. (1930). The good agreement between this graph and that of 1924 based on Stockwell's formulas is an important check. In 1930 and 1938 Milankovitch presented the calculated secular fluctuations of the radiation during the winter as well as during the summer half-year on latitudes 5°, 15°, 25°, 35°, 45°, 55°, 65°, and 75° N. and S. for the last six hundred thousand years (p. 146). While the amount of the annual solar radiation has fluctuated little through the ages, those of the caloric summer and winter half-years have varied considerably (pp. 145–49). Low summer solar radiation is accompanied by high winter radiation, and high summer insolation by low winter radiation.

As stated, the equable oceanic ages of low summer and high winter radiation were already associated by Köppen in 1924 with the ages of ice expansion. This correlation was confirmed by the German climatologist Walter Wundt, who found that a reduction of the annual temperature range by 1° C. depresses the snow line by an amount of 78 meters (p. 152), and by Milankovitch, who showed that the height of the modern snow line in the different latitudes closely correlates with the summer radiation (pp. 150–51). With cooler summers the melting of snow and ice decreased, and the snow line fell; and with milder winters the snowfall increased because of its being greatest near the freezing temperature, and the zone of maximum snowfall rose in altitude. This latter rise, together with the greater descent of the snow line, made the two levels approach each other or coincide, augmenting the snow available for glaciers (pp. 152–54). The magnitude of the climatic effects of the calculated fluctuations of the solar radiation is difficult to assess, but Zeuner modestly assumes that they lowered the snow line by 200–400 meters (pp. 152, 156). Of course, mountains are a primary condition for the development of glaciers in middle and low latitudes.

Thus the mountains and the primary effects of the low summer and high winter radiation were insufficient to produce the large Pleistocene ice sheets. However, the glaciers and ice caps that did form further modified the climate in various ways. First and foremost, they caused an increase in the percentage of the solar radi-

ation which was reflected, for the albedo of snow and ice is about 80 per cent, while that of fields, pastures, forests, and lakes is, at most, 10 per cent. This self-increasing capacity of ice sheets has been especially investigated and stressed by Wundt, who has calculated that the increased reflection during the maxima of the Pleistocene glaciations caused a temperature lowering on the Northern Hemisphere of over 4° C. (p. 156). This corresponds to a depression of the snow line of 600 meters, which is to be added to the original depression of a few hundred meters.

The ice sheets also brought about great changes in the air circulation: Glacial anticyclones formed and induced anticyclonic east winds, and the traveling cyclones were forced to take more southerly courses (pp. 156–58).

Zeuner concludes that the Pleistocene glaciations and associated climatic conditions in Europe are adequately explained by the principal summer minima and winter maxima of the solar radiation and by the supplementary effects of glaciers and ice sheets as they developed (p. 159). However, the Ice Age is not explained; for, although similar summer minima and winter maxima of insolation must have occurred periodically throughout the ages, the next preceding continental glaciation occurred in the Permian, two hundred million years ago (p. 161). The Pleistocene Ice Age apparently required some exceptional conditions favorable for glaciation; and these might have been, Zeuner believes, a Tertiary-Quaternary lowering of the sea-level, probably caused by sinking of the ocean floors and uplift of parts of the continents (p. 165).

Milankovitch's graph of the summer insolation agrees well with the geological sequences in Europe, especially those worked out by Penck and Brückner, Eberl, and Soergel (pp. 167, 170). The first extensive glaciation may have begun six hundred thousand years ago, and this date is taken by Zeuner as the demarcation line between the Tertiary and the Quaternary (p. 175).

Extension of the absolute astronomic chronology beyond the northern temperate zone meets difficulties, but Zeuner applies it for the Last Interglacial and the Last Glacial to the Mediterranean region (pp. 176–207).

Since the secular course of the summerly solar radiation is determined in high latitudes mainly by the inclination of the earth's axis, in low latitudes largely by the longitude of the perihelion and the eccentricity of the earth's orbit,[2] it changes gradually from a periodicity of forty thousand years near the poles to one of twenty-one thousand near the equator (pp. 149, 216). The main radiation minima and maxima of high latitudes occur on the entire Northern Hemisphere, but additional ones appear in mid-latitudes and grow in prominence toward the equator. It is unknown whether or not the radiation changes can explain the geologic and climatic history of the tropical zone, for their effects on the tropical climates have not been studied (pp. 217, 219). As a consequence, the Pleistocene history of tropical Africa can be correlated with that of Europe only on a general paleontological basis (pp. 208, 215).

Since the effects of the inclination of the earth's axis are identical on the Northern and the Southern Hemisphere, but those of the longitude of the perihelion and of the eccentricity are reversed, and since, as just stated, the relative influence of these factors varies with latitude, the secular course of the summer radiation in the two polar hemispheres was almost identical in high latitudes, almost reversed in low latitudes, and different in mid-latitudes.[3] In the latitudes of the main Pleistocene glaciations the culminations of the summer minima in the Northern and Southern hemispheres were offset in time at most ten thousand years, which would have made long-lasting glaciations largely contemporaneous. The climatic effects of the fluctuating summer radiation on the Southern Hemisphere are little known (p. 222).

Chapter ix, pages 225–52, deals with the Pleistocene fluctuations of the sea-level and the world-wide extension of the absolute chronology. Melting of the existing ice caps and glaciers would raise sea-level 40 to 60 meters, so that any higher abandoned shorelines must involve at least some vertical crustal movements (p. 225). In different parts of the world there are five high Quaternary shorelines or sea-levels between which there must have been relatively low ones (p. 247). The high sea-levels must be interglacial, the low ones glacial, as long held by glacialists (p. 248). The height of the old terraces decreases progressively from the oldest at 100 meters to the youngest at 7.5 meters (p. 246), a fact attributed by Zeuner to a continuous slow sinking of the sea-level (p. 248). The principal terraces and erosion channels are described, correlated, and dated (pp. 246, 252). The high sea-levels are particularly important

[2] Milutin Milankovitch, "Astronomische Mittel zur Erforschung der erdgeschichtlichen Klimate," *Handb. der Geophys.*, Band 9 (1938), pp. 593–698; see p. 661.

[3] *Ibid.*, pp. 660 and 661.

for a world-wide correlation and, Zeuner hopes, for extension of the astronomical chronology over the globe.

The concluding chapter, pages 253–78, treats the evolution of the mammals and of British land and fresh-water mollusks during the Quaternary. The composition of the faunas changed slowly by intrusion of newcomers and rapidly by a large-scale shifting with the environment (p. 256). The environment changed as the climate varied in respect to general temperature, seasonal contrast, and precipitation. In an oceanic climate the succession was from temperate forest through subarctic forest to tundra; in a continental climate from warm-continental steppe through loess steppe to tundra. However, the animals adjusted themselves very slowly, for faunas fully typical of the mentioned environments occur first in the Upper Pleistocene, that is, since the beginning of the Last Interglacial (p. 255). The most striking change in the mammal fauna was the increase of the cold component, which suggests a gradual adaptation to low temperatures (p. 274). This was brought about by migration and phylogenetic evolution. Several lineages of ascent are given. Since the earliest and the latest forms in a lineage are distinct species, it appears that half a million years were required for the evolution of a new species (pp. 277, 278).

Foremost among the many problems in this outstanding book is the absolute chronology of the Pleistocene. The modern hypothesis of the Pleistocene Ice Age employing the secular changes of the earth's position relative to the sun—which may be called the Milankovitch-Köppen-Wundt hypothesis—contains three main steps, viz.:

1. The Ice Age was made possible by a preglacial development of the most favorable conditions for glaciation since the Permian, and these conditions still prevail. The favorable changes included uplift of extensive and high mountain ranges in which glaciers could form, rise of continental shelves and submarine ridges so that these partly or practically excluded warm ocean currents from high latitudes, and lowering of the sea-level. The changes caused a temperature lowering.

2. Glaciers grew into ice fields and initial ice sheets, i.e., *extensive* glaciation was *started* by the climatic conditions induced by the extreme minima of the summer radiation and the associated maxima of the winter radiation, which were caused by certain, periodically recurring, positions of the earth in relation to the sun.

This was the most critical stage, for glaciers had to grow into effective reflectors of the solar heat, into small self-perpetuating ice sheets during the ten thousand to twelve thousand years that the periods of low summer and high winter insolation lasted. It thus depended on several factors and conditions whether or not the local glaciation developed into a continental glaciation. Some factors were: extent of glaciers and of sea ice at the onset of low summer insolation; the degree of change in the solar radiation; the time of beginning effectiveness of the expansion of the sea ice, of the reduction of the Gulf Stream water entering the Norwegian Sea, and of the reduction of the heat transport to high latitudes by the migratory cyclones; and the intensity of the summer radiation during the succeeding cycle. Even though the present-day ice recession suggests that glaciers are very sensitive to climatic change, a primary decrease of the annual temperature range by several degrees centigrade may have been necessary (cf. pp. 147, 149, 152). Thus, not every minimum of summer insolation, especially not the first one following upon an interglacial, need have been, or may have been, represented by ice sheets. For instance, there is no convincing evidence that ice sheets formed upon the radiation minimum 115,000 B.P. (before the present), though Zeuner so believes (pp. 53, 169). In America the Iowan glacial maximum, dated by G. F. Kay at 55,000 B.P., followed the radiation minimum of 72,000 B.P.; and the Mankato glacial culmination, as estimated at 25,000 B.P., approximately coincided with the radiation minimum of some twenty-three thousand years ago. Furthermore, since the double minima of summer radiation which are correlated with the first three large glaciations are spaced only forty .thousand to forty-three thousand years, since the formation of ice sheets may have required more or less than twenty-five thousand years, and since there must have been a very considerable lag of the growth and shrinking of the ice sheets (pp. 160, 168), the continental glaciations and the sea-level fluctuations need not have been doubled.

3. The ice fields expanded into vast ice sheets with the help of secondary effects, especially cooling by the increasing reflection of the sun's heat (Wundt). This occurred at least partly during somewhat stronger summer insolation than is received today. The growing ice sheets caused the development of glacial anticyclones and anticyclonic winds and deflected the migratory cyclones. The pack ice expanded to fill the

North Atlantic southward perhaps to Cornwall and the banks off Nova Scotia.[4] Most drifting ice was carried westward on the north side of the mean storm tracks, but some reached the southerly belt of westerly winds and was transported by the anticyclonic whirl toward southwestern Europe and then south along the coast of Portugal. The Gulf Stream was deflected southward with the cyclone tracts until even its northeasternmost branches, now entering the Norwegian Sea, were incorporated in the clockwise whirl. The glacial anticyclones and the southerly position of the Icelandic and Aleutian barometric lows favored low-index circulation (weak westerlies in the middle latitudes,[5] during which strong cold waves penetrated exceptionally far south, but warm tropical air was carried only moderately far north. Therefore, while there is at present an enormous winterly importation of heat to Europe by the migratory cyclones and the Gulf Stream—the January temperature is on the Norwegian northwest coast 20° C. (36° F.), in central Europe 8° C. (14.4° F.) above the means for the latitudes—there was then little if any heat importation, and as a consequence the winter temperature may at the time of the glacial culminations have been low enough for the observed glaciations. Also the summer temperature, which was reduced by the ice sheets, by drifting ice and chilly waters of the Atlantic, and by fogs, may ultimately have become sufficiently low to account for the Pleistocene glaciers in Ireland, Portugal, and southwestern Yugoslavia which Penck[6] believed required a primary reduction of the annual temperature of at least 8° C. (14.4° F.).

These and other effects of the ice sheets assisted in spreading the glaciations and in creating the climatic conditions of the glacials.

The Milankovitch-Köppen-Wundt hypothesis of the Pleistocene Ice Age has a physical basis throughout and does not make unverifiable assumptions of, for instance, a lowered or fluctuating solar constant. Extensive mountain building and a general cooling preceded the Ice Age. Secular changes in the earth's solar position occur and have been calculated and dated for the last one million years; and they influence the earth's insolation and climate. The extreme minima of summer and maxima of winter insolation must have brought about at least a notable increase of glaciation; and the approximate coincidence of their dates and the estimated ages of the glacials suggests a causal relationship.[7] Secondary effects of ice sheets, some of them chain effects, are even at this early stage of their study known to have played an important part. Several independent as well as interrelated factors and causes rather than a few or a single one are in accord with the complexity of the climatic and geologic conditions and events of the Ice Age. For instance, widespread glaciation required primarily lower temperature in the British Isles and in the Scandinavian mountains and heavier snowfall in the Labrador Peninsula; the last great (Mankato, Tioga) ice advance between the Pacific and the Mississippi River coincided with ice retreat in the northeastern states. This composite hypothesis has changed in important respects and developed along new lines since first advanced by Köppen in 1924, and it is in the stage of growth. It opens a wide and promising field of research.

Zeuner's endeavors to correlate the geological history or relative chronology of the Pleistocene with the history of the fluctuating summer insolation therefore deserve serious and open-minded consideration. The probability that not every extreme minimum of summer insolation need have led to a continental glaciation and our inability as yet to appraise the several secondary effects of the ice sheets and the lag of expansion and of shrinking of the ice advise verification of details. Considerable study lies ahead.

Zeuner has rendered a great service by his able investigation and treatment of these exceptionally difficult fundamental problems of the Ice Age.

ERNST ANTEVS

Postglacial Forest Succession, Climate, and Chronology in the Pacific Northwest. By HENRY P. HANSEN. Philadelphia: Transactions American Philosophical Society, Vol. XXXVII (Part I), 1947. Pp. 130; figs. 114. $2.25.

[4] See also G. C. Simpson, "World Climate during the Quaternary Period," *Quart. Jour. Roy. Met. Soc.*, Vol. LX (1934), pp. 424–78; see pp. 431–37; *ibid.*, "Ice Ages," *Nature*, Vol. CXLI, No. 3570 (1938), pp. 591–98; *ibid.*, *Smithsonian Rept. for 1938*, pp. 289–302.

[5] C. G. Rossby, "The Scientific Basis of Modern Meteorology," *U.S. Dept. Agric. Yearbook 1941*, pp. 599–655; see pp. 621–31.

[6] Pp. 333–35 and 347–48 of ftn. 1.

[7] Albrecht Penck and Eduard Brückner, *Die Alpen im Eiszeitalter* (Leipzig, 1909), Vol. III, p. 1168; and G. F. Kay, "Classification and Duration of the Pleistocene Period," *Bull. Geol. Soc. Amer.*, Vol. XLII (1931), pp. 425–66.

Since Professor Hansen of Oregon State College began his western plant pollen studies in 1935, he has examined most peat bogs and many other pollen-bearing sediments in Oregon, Washington, and northern Idaho, has described the pollen profiles, and has discussed their significance in a long series of papers. Now he has brought together his pollen data from some seventy sedimentary columns in a single volume in which he treats the methodic, botanical, ecological, climatic, and geochronologic problems and draws important conclusions.

This review covers the chapters on geology, past climate, and geochronology (pp. 8–19, 33–35, 108–22). Other subjects in the monograph are: history and theory of pollen analysis, pollen-bearing profiles of the Pacific Northwest, rate of organic deposition, methods and technique, climate of the Pacific Northwest, forests of the Pacific Northwest, characteristics of Pacific Northwest forest trees, and postglacial vegetation history.

Most of the lake basins in which peat and other pollen-bearing deposits have accumulated in the Pacific Northwest were of direct glacial origin. Others were caused by damming of glacial streams, erosion by such streams, eustatic changes of sea level, and ponding of tributary streams by aggradation of the main valley. Still other basins were formed by landsliding, faulting, ponding by lava flows and beaver dams, and by severing of oxbows (pp. 8–19).

Layers of pumice and volcanic ash in the columns have served as excellent correlation markers (pp. 3, 37, 38, 114, 118, 119).

The culmination of the last continental glaciation, the Mankato, is given the usual age of 25,000 years, and the deposition of the pollen-bearing sediments resting on glacial drift is assumed to have begun some 18,000–20,000 years ago. The age of the pollen records has been arbitrarily called the Postglacial. On changes in the forest composition, shown for the individual regions in a table, this Postglacial has been divided into these four climatic periods (pp. 4, 35, 113–151:

Year	Period
0	the present
4,000	IV, cooler and moister
8,000	III, maximum warmth and dryness
15,000	II, increasing warmth and dryness
18,000–20,000	I, cool and moist

During the last glacial maximum the snow line in the Cascades was depressed some 1,400 meters (4,600 feet), according to Fritz Klute;[1] and the maxima of lodgepole pine and western white pine in the bottom layers of the sediments indicate cooler and moister climate than at any later time, but low percentage of pollen of boreal species suggests absence of tundra (p. 113).

A distinctly warmer age a few thousand years ago is recorded in several regions of the north temperate zone, and it has been dated in Sweden and Denmark at about 8000–4000 B.P. (before the present). The terminal date is also calculated from the historical salt contents in Owens Lake, California, and in Abert and Summer lakes, Oregon. This warm age was also dry in the western United States and is recorded not only by the forests of the Northwest but by deglaciation in the western mountains and by arroyo cutting and wind erosion in the Southwest (pp. 4, 111, 119).

A return to cooler and moister climate some 4,000 years ago is indicated in the western states by expansion of moisture-loving trees, rebirth of lakes and glaciers, filling of arroyos, and anchoring of dunes (pp. 119, 120).

Hansen consequently reads in his pollen profiles the same main climatic trends, the same general climatic history as has been deduced from various geological evidence in the western United States. Although no subordinate climatic variations seem to be recorded by the pollen profiles of the Northwest, there were surely in most regions additional fluctuations of several orders, that is, of different duration, amplitude, and geographic extent. Paul Sears and E. S. Deevey find that, for instance, in the northeastern United States the progressive dryness after the departure of the ice was broken, and a moist age intervened next before the age of maximum warmth and dryness (pp. 110–12). In southeastern Arizona the age of general deposition which began about four thousand years ago was temporarily interrupted by drought-induced erosion about the time of Christ and about A.D. 1300. Natural erosion would perhaps have set in again during the dry 1930's, had not overgrazing started artificial erosion in 1883. As knowledge increases, more climatic fluctuations and regional differences will be recognized, the latter mainly in precipitation and moisture.

It was to enable long-distance correlations in

[1] "Die Bedeutung der Depression der Schneegrenze für eiszeitliche Probleme," *Zeitschr. f. Gletscherkunde*, Vol. XVI (1928), pp. 70–93; see p. 75.

spite of such subordinate regional differences that Lennart von Post and I independently singled out the predominant and most widespread climatic change for time division of the Postglacial, namely, the rise, culmination, and fall of the temperature (pp. 109–11). In making correlations, it must be realized that peripheral regions were released from the ice sheets tens of thousands of years before central ones and that all the regions have had remarkably similar climatic histories despite the unequal lengths of time involved. Most correlations have to be made on a differentially sliding time scale (pp. 111, 112).

Hansen's monograph is the most comprehensive study of pollen profiles and related problems carried out in North America; it is ably done and beautifully illustrated.

ERNST ANTEVS

Charles Darwin and the Voyage of the Beagle. Edited with an Introduction by NORA BARLOW. Philosophical Library, 1946. Pp. 279; frontispiece; pls. 15; and map. $3.75.

This volume, compiled by a granddaughter, begins with a brief biography of Darwin's life to the age of twenty-one when, in 1831, he embarked on H.M.S. "Beagle," a sailing vessel of two hundred and forty-two tons and ten guns, as naturalist. At that time he appears to have been somewhat neurotic, totally lacking in ambition and completely dominated by the overpowering personality of his well-to-do father, who gave reluctant permission for the son to go adventuring only upon the intercession of an uncle, who argued that "the pursuit of Natural History, though certainly not professional, is very suitable to a clergyman."

Darwin's formal education seems to have been completely wasted—the classics and ancient history, medicine at the University of Edinburgh, and preparation for the clergy at Cambridge. Much of his spare time, however, had been devoted to the hobby of "Natural History" which was his only serious interest, but even this was not permitted to interfere with such pleasures as partridge shooting on the estate of relatives. His appointment (he received no salary and was obliged to pay his own expenses) was secured through the intercession of friends and was also influenced by the prejudice of the vessel's captain who refused to "take anyone, however good a naturalist, who was not recommended to him, likewise, as a gentleman." Altogether, young Darwin appears to have been a not too promising choice, and he certainly was singularly unprepared for such an undertaking.

The main portion of the book is divided into two nearly equal parts. The first consists of thirty-eight letters mostly written to his sisters in England and mainly unpublished previously. The letters are largely devoted to personal matters and briefly describe some of his impressions and activities principally in South America. They are of most interest because here and there they give hints of his gradual maturity, increasing self-reliance, and the steady growth of his interest in natural history and especially geology. Obviously, he returned home after five years a different man with a serious purpose in life that previously had been lacking.

The second part consists of a selection of more or less indiscriminate extracts from twenty-four field notebooks in which Darwin jotted down his observations on all subjects. These notebooks have not been published, but they furnished the basis for his *Journal*, which appeared in several editions, and the three-volume account of his geological studies. The extracts from these notes are connected by comments and interpretations by the compiler. Many of them are so abbreviated as to be almost unintelligable to the reader, and the discontinuity of the subject matter is so great as to suggest that Darwin's observations were all made in the most hit-or-miss fashion. The Introduction states that the notes are largely geological and that from half to nearly nine-tenths of the entries in different books are devoted to this subject. Perhaps because the compiler had little or no knowledge of geology, these parts have been slighted in the selection, and thus what may have been his most significant observations have been omitted.

This book is neither a very interesting nor important contribution to Darwiniana, and similar writings of a lesser man could never find a publisher. It contains little that is not better presented in Darwin's own volumes or in the previously published Darwin letters. Illustrations consist of fifteen plates of rather poorly reproduced drawings, a portrait of Darwin and one of his sisters as children, and a map showing the route travelled by the "Beagle."

J. MARVIN WELLER